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Isopentylcarboxanilides

The present invention relates to novel isopentylcarboxanilides, to a plurality of processes for their preparation and to their use for controlling unwanted microorganisms.

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It is already known that numerous carboxanilides have fungicide properties (cf., for example, WO 02/059086, WO 00/09482, EP-A 0 824 099, EP-A 0 755 927, EP-A 0 589 301, EP-A 0 545 099, JP 11-335364, JP 10-310577 and JP 10-251240). Known are, for example, 1-methyl-N-[2-(3methylbutyl)phenyl]-3-(trifluoromethyl)-1H-pyrazole-4-carboxamide (from EP-A 0 824 099) and 2,5-dimethyl-N-[3-(3-methylbutyl)phenyl]-3-furamide from (EP-A 0 755 927). The activity of these compounds is good; however, it is sometimes unsatisfactory at low application rates.

This invention now provides novel isopentylcarboxanilides of the formula (I)

15 in which

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L represents
$$\stackrel{}{\downarrow_2}$$
 $\stackrel{}{\downarrow_3}$ $\stackrel{}{\downarrow_4}$ $\stackrel{}{\downarrow_4$

where the bond labelled with * is attached to the amide, whereas the bond labelled with # is attached to the alkyl side chain,

represents hydrogen,, C₁-C₈-alkyl, C₁-C₆-alkylsulphinyl, C₁-C₆-alkylsulphonyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl; C₁-C₆-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄-haloalkylsulphinyl, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl-C₁-C₃alkyl, $(C_1-C_3-alkyl)$ carbonyl- $C_1-C_3-alkyl$, $(C_1-C_3-alkoy)$ carbonyl- $C_1-C_3-alkyl$; halo- $(C_1-C_3-alkyl)$ alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms;

 $(C_1-C_8-alkyl)$ carbonyl, $(C_1-C_8-alkoxy)$ carbonyl, $(C_1-C_4-alkoxy-C_1-C_4-alkyl)$ carbonyl, $(C_3-C_8-alkyl)$ cycloalkyl)carbonyl; (C₁-C₆-haloalkyl)carbonyl, (C₁-C₆-haloalkoxy)carbonyl, (halo-C₁-C₄alkoxy-C₁-C₄-alkyl)carbonyl, (C₃-C₈-halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O)R⁴, -CONR⁵R⁶ or -CH₂NR⁷R⁸,

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 R^1

R² represents hydrogen, fluorine, chlorine, methyl or trifluoromethyl,

R³ represents hydrogen, halogen, C₁-C₈-alkyl, C₁-C₈-haloalkyl,

R⁴ represents hydrogen, C₁-C₈-alkyl, C₁-C₈-alkoxy, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl; C₁-C₆-haloalkyl, C₁-C₆-haloalkoxy, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms,

R⁵ and R⁶ independently of one another each represent hydrogen, C₁-C₈-alkyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl; C₁-C₈-haloalkyl, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms,

R⁵ and R⁶ furthermore together with the nitrogen atom to which they are attached form a saturated heterocycle having 5 to 8 ring atoms which is optionally mono- or polysubstituted by identical or different substituents from the group consisting of halogen and C₁-C₄-alkyl, where the heterocycle may contain 1 or 2 further non-adjacent heteroatoms from the group consisting of oxygen, sulphur and NR⁹,

R⁷ and R⁸ independently of one another represent hydrogen, C₁-C₈-alkyl, C₃-C₈-cycloalkyl; C₁-C₈-haloalkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms.

R⁷ and R⁸ furthermore together with the nitrogen atom to which they are attached form a saturated heterocycle having 5 to 8 ring members which is optionally mono- or polysubstituted by identical or different substituents from the group consisting of halogen and C₁-C₄-alkyl, where the heterocycle may contain 1 or 2 further non-adjacent heteroatoms from the group consisting of oxygen, sulphur and NR⁹,

R⁹ represents hydrogen or C₁-C₆-alkyl,

A represents the radical of the formula (A1)

$$R^{10}$$
 N
 R^{11}
 R^{12}
(A1), in which

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R¹⁰ represents hydrogen, hydroxyl, formyl, cyano, halogen, nitro, C₁-C₄-alkyl, C₁-C₄-alkyl, C₁-C₄-alkylthio, C₃-C₆-cycloalkyl, C₁-C₄-haloalkyl, C₁-C₄-haloalkoxy or C₁-C₄-haloalkylthio having in each case 1 to 5 halogen atoms, aminocarbonyl or aminocarbonyl -C₁-C₄-alkyl,

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R¹¹ represents hydrogen, halogen, cyano, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-haloalkyl or C₁-C₄-haloalkylthio having in each case 1 to 5 halogen atoms, and

R¹² represents hydrogen, C₁-C₄-alkyl, hydroxy-C₁-C₄-alkyl, C₂-C₆-alkenyl, C₃-C₆-cyclo-alkyl, C₁-C₄-alkylthio-C₁-C₄-alkyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₁-C₄-haloalkyl, C₁-C₄-alkyl, C₁-C

haloalkylthio-C₁-C₄-alkyl, C₁-C₄-haloalkoxy-C₁-C₄-alkyl having in each case 1 to 5 halogen atoms, or represents phenyl,

with the proviso that R^{10} does not represent iodine if R^{11} represents hydrogen and with the proviso that R^{10} does not represent trifluoromethyl or difluoromethyl if R^3 and R^{11} represent hydrogen and R^{12} represents methyl,

· or

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represents the radical (A2)

 R^{13} and R^{14} independently of one another represent hydrogen, halogen, C_1 - C_4 -alkyl or C_1 - C_4 -haloalkyl having 1 to 5 halogen atoms and

 R^{15} represents halogen, cyano or C_1 - C_4 -alkyl, or C_1 - C_4 -haloalkyl or C_1 - C_4 -haloalkoxy having in each case 1 to 5 halogen atoms,

or

A represents the radical of the formula (A3)

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 R^{16} and R^{17} independently of one another represents hydrogen, halogen, C_1 - C_4 -alkyl or C_1 - C_4 -haloalkyl having 1 to 5 halogen atoms and

R¹⁸ represents hydrogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

20 A represents the radical of the formula (A4)

R¹⁹ represents hydrogen, halogen, hydroxyl, cyano, C₁-C₆-alkyl, C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio having in each case 1 to 5 halogen atoms,

or

25 A represents the radical of the formula (A5)

$$\mathbb{R}^{21}$$
 \mathbb{N} \mathbb{R}^{20} (A5), in which

R²⁰ represents halogen, hydroxyl, cyano, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio or C₁-C₄-haloalkoxy having in each case 1 to 5 halogen atoms and

R²¹ represents hydrogen, halogen, cyano, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-haloalkyl, C₁-C₄-haloalkoxy having in each case 1 to 5 halogen atoms, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl,

or

5 A represents the radical of the formula (A6)

$$\bigcirc$$
 CH₃ (A6),

or

A represents the radical of the formula (A7)

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R²² represents C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or A

represents the radical of the formula (A8)

R²³ represents C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

15 or

A represents the radical of the formula (A9)

$$\mathbb{R}^{25}$$
 (A9), in which

R²⁴ and R²⁵ independently of one another represents hydrogen, halogen, amino, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms and

20 R^{26} represents hydrogen, C_1 - C_4 -alkyl or C_1 - C_4 -haloalkyl having 1 to 5 halogen atoms, with the proviso that R^{24} and R^{26} do not simultaneously represent methyl if R^{25} represents hydrogen,

or

A represents the radical of the formula (A10)

$$R^{28}$$
 R^{29} (A10) in which

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R²⁷ and R²⁸ independently of one another represent hydrogen, halogen, amino, nitro, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms and

R²⁹ represents halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

A represents the radical of the formula (A11)

$$R^{30}$$
 (A11) in which

R³⁰ represents hydrogen, halogen, amino, C₁-C₄-alkylamino, di-(C₁-C₄-alkyl)amino, cyano, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms and

R³¹ represents halogen, hydroxyl, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₃-C₆-cycloalkyl, C₁-C₄-haloalkyl or C₁-C₄-haloalkoxy having in each case 1 to 5 halogen atoms,

with the proviso that R³¹ does not represent trifluoromethyl, difluoromethyl or methyl if R³ represents hydrogen and R³⁰ represents methyl,

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or

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A represents the radical of the formula (A12)

$$R^{32}$$
 (A12) in which

 R^{32} represents hydrogen, halogen, amino, C_1 - C_4 -alkylamino, di- $(C_1$ - C_4 -alkyl)amino, cyano, C_1 - C_4 -alkyl or C_1 - C_4 -haloalkyl having 1 to 5 halogen atoms and

R³³ represents halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

A represents the radical of the formula (A13)

$$R^{34}$$
 (A13) in which

 R^{34}

represents hydrogen or C₁-C₄-alkyl and

R³⁵ represents halogen or C₁-C₄-alkyl,

or

A represents the radical of the formula (A14)

$$(A14)$$
 in which

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R³⁶ represents hydrogen, halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

A represents the radical of the formula (A15)

R³⁷ represents halogen, hydroxyl, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio or C₁-C₄-haloalkoxy having in each case 1 to 5 halogen atoms,

5 or

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A represents the radical of the formula (A16)

$$R^{40}$$
 R^{39}
 R^{39}
 R^{41}
 R^{38}
(A16) in which

R³⁸ represents hydrogen, cyano, C₁-C₄-alkyl, C₁-C₄-haloalkyl having 1 to 5 halogen atoms, C₁-C₄-alkoxy-C₁-C₄-alkyl, hydroxy-C₁-C₄-alkyl, C₁-C₄-alkylsulphonyl, di(C₁-C₄-alkyl)aminosulphonyl, C₁-C₆-alkylcarbonyl or in each case optionally substituted phenylsulphonyl or benzoyl,

R³⁹ represents hydrogen, halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

R⁴⁰ represents hydrogen, halogen, cyano, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

R⁴¹ represents hydrogen, halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

with the proviso that R⁴⁰ does not represent trifluoromethyl,

or

20 A represents the radical of the formula (A17)

R⁴² represents C₁-C₄-alkyl.

If appropriate, the compounds according to the invention can be present as mixtures of various possible isomeric forms, in particular of stereoisomers, such as, for example, E and Z, threo and erythro, and also optical isomers, and, if appropriate, also of tautomers. What is claimed are both the E and the Z isomers, and also the threo and erythro and the optical isomers, any mixtures of these isomers and the possible tautomeric forms.

Furthermore, it has been found that isopentylcarboxanilides of the formula (I) are obtained when

a) carboxylic acid derivatives of the formula (II)

$$\begin{array}{ccc}
 & & & & \\
 & & & \\
A & & & \\
X^1 & & & \\
\end{array} (II),$$

in which

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or

A is as defined above and

X¹ represents halogen or hydroxyl,

are reacted with an aniline derivative of the formula (III)

$$\begin{array}{c|c}
HN & L \\
R^1 & R^3 \\
H_3C & CH_3
\end{array}$$
(III)

in which L, R¹ and R³ are as defined above,

if appropriate in the presence of a catalyst, if appropriate in the presence of a condensing agent, if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent,

b) isopentylcarboxanilides of the formula (I-a)

in which

L, A and R³ are as defined above are reacted with halides of the formula (IV)

$$R^{1-A} X^2$$
 (IV)

in which

20 X² represents chlorine, bromine or iodine,

R^{1-A} represents C₁-C₈-alkyl, C₁-C₆-alkylsulphinyl, C₁-C₆-alkylsulphonyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl; C₁-C₆-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl; halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl in each case having 1 to 13 fluorine, chlorine and/or bromine atoms; (C₁-C₈-alkyl)carbonyl, (C₁-C₈-alkoxy)carbonyl, (C₁-C₆-haloalkyl)carbonyl, (C₁-C₆-haloalkoxy)carbonyl, (C₁-C₆-haloalkoxy)carbonyl,

(halo- C_1 - C_4 -alkoxy- C_1 - C_4 -alkyl)carbonyl, (C_3 - C_8 -halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O) R^4 , CON R^5R^6 or - $CH_2NR^7R^8$,

where R⁴, R⁵, R⁶, R⁷ and R⁸ are as defined above,

in the presence of a base and in the presence of a diluent,

or

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c) isopentone derivatives of the formula (V)

in which

 R^1 , R^2 , R^3 and A are as defined above,

are reacted with hydrazine (or hydrazine hydrate) in the presence of a base and, if appropriate, in the presence of a diluent,

or

d) isopentene derivatives of the formula (VI)

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in which R1, R2, R3 and A are as defined above,

are hydrogenated, if appropriate in the presence of a diluent and if appropriate in the presence of a catalyst,

or

20 e) isopentyne derivatives of the formula (VII)

in which R1, R2, R3 and A are as defined above,

are hydrogenated, if appropriate in the presence of a diluent and if appropriate in the presence of a catalyst.

Finally, it has been found that the novel isopentylcarboxanilides of the formula (I) have very good microbicidal properties and can be used for controlling unwanted microorganisms both in crop protection and in the protection of materials.

- 5 The formula (I) provides a general definition of the isopentylcarboxanilides according to the invention. Preferred radical definitions of the formulae shown above and below are given below. These definitions apply both to the end products of the formula (I) and likewise to all intermediates.
- L preferably represents L-1 where R² may in each case have the general, preferred, particularly preferred, very particularly preferred or especially preferred meanings.
 - L furthermore preferably represents L-2.
 - L furthermore preferably represents L-3.
 - L furthermore preferably represents L-4.
- L <u>particularly preferably</u> represents L-1, where R² may in each case have the general, preferred, particularly preferred, very particularly preferred or especially preferred meanings.
 - L furthermore <u>particularly preferably</u> represents L-2.
 - L <u>very particularly preferably</u> represents L-1 where R² in each case have the general, preferred, particularly preferred, very particularly preferred or especially particularly preferred meanings.

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preferably represents hydrogen, C₁-C₆-alkyl, C₁-C₄-alkylsulphinyl, C₁-C₄-alkylsulphonyl, C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-cycloalkyl; C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄-haloalkylsulphinyl, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl; halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms;

(C₁-C₆-alkyl)carbonyl, (C₁-C₄-alkoxy)carbonyl, (C₁-C₃-alkoxy-C₁-C₃-alkyl)carbonyl, (C₃-C₆-cycloalkyl)carbonyl; (C₁-C₄-haloalkyl)carbonyl, (C₁-C₄-haloalkoxy)carbonyl, (halo-C₁-C₃-alkyl)carbonyl, (C₃-C₆-halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O)R⁴, -CONR⁵R⁶ or -CH₂NR⁷R⁸.

R¹ <u>particularly preferably</u> represents hydrogen, methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl, pentyl or hexyl, methylsulphinyl, ethylsulphinyl, n- or isopropylsulphinyl, n-, iso-, sec- or tert-butylsulphinyl, methylsulphonyl, ethylsulphonyl, n- or isopropylsulphonyl, n-, iso-, sec- or tert-butylsulphonyl, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxymethyl, cyclopropyl, cyclopentyl, cyclohexyl, trifluoromethyl, trichloromethyl, trifluoroethyl,

difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethylsulphinyl, trifluoromethylsulphonyl, trifluoromethoxymethyl; formyl, -CH2-CHO, -(CH2)2-CHO, -CH₂-CO-CH₃, -CH₂-CO-CH₂CH₃, -CH₂-CO-CH(CH₃)₂, -(CH₂)₂-CO-CH₃, -(CH₂)₂-CO-CH₂CH₃, -(CH₂)₂-CO-CH(CH₃)₂,-CH₂-CO₂CH₃, -CH₂-CO₂CH₂CH₃, $-CH_2-CO_2CH(CH_3)_2$, -(CH₂)₂-CO₂CH₃, -(CH₂)₂-CO₂CH₂CH₃, -(CH₂)₂-CO₂CH(CH₃)₂, -CH₂-CO-CF₃, -CH₂-CO-CCl₃, -CH₂-CO-CH₂CF₃, -CH₂-CO-CH₂CCl₃, -(CH₂)₂-CO-CH₂CF₃, -(CH₂)₂-CO-CH₂CCl₃, -CH₂-CO₂CH₂CF₃, -CH₂-CO₂CF₂CF₃, -CH₂-CO₂CH₂CCl₃, $-CH_2-CO_2CCl_2CCl_3$, $-(CH_2)_2-CO_2CH_2CF_3$, $-(CH_2)_2-CO_2CF_2CF_3$, -(CH₂)₂-CO₂CH₂CCl₃, -(CH₂)₂-CO₂CCl₂CCl₃;

- methylcarbonyl, ethylcarbonyl, n-propylcarbonyl, isopropylcarbonyl, tert-butylcarbonyl, methoxycarbonyl, ethoxycarbonyl, tert-butoxycarbonyl, cyclopropylcarbonyl; trifluoromethylcarbonyl, trifluoromethoxycarbonyl, or -C(=O)C(=O)R⁵, -CONR⁶R⁷ or -CH₂NR⁸R⁹.
- R¹ <u>very particularly preferably</u> represents hydrogen, methyl, methoxymethyl, formyl, -CH₂-CHO, -(CH₂)₂-CHO, -CH₂-CO-CH₃, -CH₂-CO-CH₂CH₃, -CH₂-CO-CH(CH₃)₂, -C(=O)CHO, -C(=O)C(=O)CH₃, -C(=O)C(=O)CH₂OCH₃, -C(=O)CO₂CH₃, -C(=O)CO₂CH₂CH₃.
 - R² <u>preferably</u> represents hydrogen.
- furthermore <u>preferably</u> represents fluorine, where fluorine is <u>particularly preferably</u> located in the 4-, 5- or 6-position, <u>very particularly preferably</u> in the 4- or 6-position, <u>especially</u> in the 4-position, of the anilide radical [cf. formula (I) above].
 - R² furthermore <u>preferably</u> represents chlorine, where chlorine is <u>particularly preferably</u> located in the 5-position of the anilide radical [cf. formula (I) above]. Chlorine is furthermore <u>particularly preferably</u> located in the 4-position of the anilide radical.
- 25 R² furthermore <u>preferably</u> represents methyl, where methyl is <u>particularly preferably</u> located in the 3-position of the anilide radical [cf. formula (I) above].
 - R² furthermore <u>preferably</u> represents trifluoromethyl, where trifluoromethyl is <u>particularly</u> <u>preferably</u> located in the 4- or 5-position of the anilide radical [cf. formula (I) above].
- R³ <u>preferably</u> represents hydrogen, fluorine, chlorine, bromine, iodine, C₁-C₆-alkyl, C₁-C₆-30 haloalkyl having 1 to 13 fluorine, chlorine and/or bromine atoms.
 - R³ particularly preferably represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, n-, isopropyl, n-, iso-, sec-, tert-butyl or C₁-C₄-haloalkyl having 1 to 9 fluorine, chlorine and/or bromine atoms.
- R³ <u>very particularly preferably</u> represents hydrogen fluorine, chlorine or methyl, ethyl or trifluoromethyl.

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- R⁴ <u>preferably</u> represents hydrogen, C₁-C₆-alkyl, C₁-C₄-alkoxy, C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-cycloalkyl; C₁-C₄-haloalkyl, C₁-C₄-haloalkoxy, halo-C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms.
- R⁴ <u>particularly preferably</u> represents hydrogen, methyl, ethyl, n- or iso-propyl, tert-butyl, methoxy, ethoxy, n- or isopropoxy, tert-butoxy, cyclopropyl; trifluoromethyl, trifluoromethoxy.
- R^5 and R^6 independently of one another <u>preferably</u> represent hydrogen, C_1 - C_6 -alkyl, C_1 - C_3 -alkoxy- C_1 - C_3 -alkyl, C_3 - C_6 -cycloalkyl; C_1 - C_4 -haloalkyl, halo- C_1 - C_3 -alkoxy- C_1 - C_3 -alkyl, C_3 - C_6 -halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms.
- 10 R⁵ and R⁶ furthermore together with the nitrogen atom to which they are attached <u>preferably</u> represent a saturated heterocycle having 5 to 8 ring atoms which is optionally mono- to tetrasubstituted by identical or different substituents from the group consisting of halogen and C₁-C₄-alkyl, where the heterocycle may contain 1 or 2 further non-adjacent heteroatoms from the group consisting of oxygen, sulphur and NR⁹.
- 15 R⁵ and R⁶ independently of one another <u>particularly preferably</u> represent hydrogen, methyl, ethyl, nor isopropyl, n-, iso-, sec- or tert-butyl, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, cyclopropyl, cyclopentyl, cyclohexyl; trifluoromethyl, trifluoromethyl, trifluoromethoxymethyl.
- R⁵ and R⁶ furthermore together with the nitrogen atom to which they are attached <u>particularly</u>

 20 <u>preferably</u> represent a saturated heterocycle from the group consisting of morpholine, thiomorpholine and piperazine which is optionally mono- to tetrasubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine and methyl, where the piperazine may be substituted on the second nitrogen atom by R9.
- R⁷ and R⁸ independently of one another <u>preferably</u> represent hydrogen, C₁-C₆-alkyl, C₃-C₆-cycloalkyl; C₁-C₄-haloalkyl, C₃-C₆-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms.
 - R⁷ and R⁸ furthermore together with the nitrogen atom to which they are attached <u>preferably</u> represent a saturated heterocycle having 5 to 8 ring atoms which is optionally mono- to tetrasubstituted by identical or different substituents from the group consisting of halogen and C₁-C₄-alkyl, where the heterocycle may contain 1 or 2 further non-adjacent heteroatoms from the group consisting of oxygen, sulphur and NR⁹.
 - R⁷ and R⁸ independently of one another <u>particularly preferably</u> represent hydrogen, methyl, ethyl, nor isopropyl, n-, iso-, sec- or tert-butyl, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, cyclopropyl, cyclopentyl, cyclohexyl; trifluoromethyl, trifluoromethyl, trifluoromethyl, trifluoromethyl,

- R⁷ and R⁸ furthermore together with the nitrogen atom to which they are attached <u>particularly</u> <u>preferably</u> represent a saturated heterocycle from the group consisting of morpholine, thiomorpholine and piperazine which is optionally mono- to tetrasubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine and methyl, where the piperazine may be substituted on the second nitrogen atom by R⁹.
- R⁹ <u>preferably</u> represents hydrogen or C₁-C₄-alkyl.
- R⁹ <u>particularly preferably</u> represents hydrogen, methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl.

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- A particularly represents one of the radicals
 A1, A2, A3, A4, A5, A6, A9, A10, A11, A12, A13, A14, A15 or A16 given above.
- A particularly preferably represents one of the radicals
 A1, A2, A4, A5, A6, A9, A11, A12, A13, A14, A15 or A16 given above.
- 15 A <u>very particularly preferably</u> represents the radical A1.
 - A furthermore <u>very particularly preferably</u> represents the radical A2.
 - A furthermore <u>very particularly preferably</u> represents the radical A4.
 - A furthermore <u>very particularly preferably</u> represents the radical A5.
 - A furthermore <u>very particularly preferably</u> represents the radical A6.
- 20 A furthermore <u>very particularly preferably</u> represents the radical A9.
 - A furthermore <u>very particularly preferably</u> represents the radical A11.
 - A furthermore <u>very particularly preferably</u> represents the radical A12.
 - A furthermore <u>very particularly preferably</u> represents the radical A13.
 - A furthermore <u>very particularly preferably</u> represents the radical A14.
- 25 A furthermore <u>very particularly preferably</u> represents the radical A16.
 - R¹⁰ preferably represents hydrogen, hydroxyl, formyl, cyano, fluorine, chlorine, bromine, iodine, methyl, ethyl, isopropyl, methoxy, ethoxy, methylthio, ethylthio, cyclopropyl, C₁-C₂-haloalkyl, C₁-C₂-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms, trifluoromethylthio, difluoromethylthio, aminocarbonyl, aminocarbonylmethyl or aminocarbonylethyl,
 - with the proviso that R¹⁰ does not represent iodine if R¹¹ represents hydrogen and with the proviso that R¹⁰ does not represent trifluoromethyl or difluoromethyl if R³ and R¹¹ represent hydrogen and R¹² represents methyl.
- 35 R¹⁰ <u>particularly preferably</u> represents hydrogen, hydroxyl, formyl, fluorine, chlorine, bromine, iodine, methyl, ethyl, isopropyl, methoxy, ethoxy, monofluoromethyl, monofluoroethyl,

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difluoromethyl, trifluoromethyl, difluorochloromethyl, trichloromethyl, dichloromethyl, pentafluoroethyl, cyclopropyl, methoxy, ethoxy, trifluoromethoxy, difluoromethoxy, trichloromethoxy, methylthio, ethylthio, trifluoromethylthio or difluoromethylthio,

with the proviso that R^{10} does not represent iodine if R^{11} represents hydrogen and with the proviso that R^{10} does not represent trifluoromethyl or difluoromethyl if R^3 and R^{11} represent hydrogen and R^{12} represents methyl.

- R¹⁰ <u>very particularly preferably</u> represents hydrogen, hydroxyl, formyl, fluorine, chlorine, bromine, iodine, methyl, ethyl, isopropyl, methoxy, cyclopropyl, monofluoromethyl, monofluoromethyl, difluoromethyl, difluoromethyl, trifluoromethyl, difluoromethyl, trichloromethyl, -CHFCH₃ or difluoromethoxy.
 - with the proviso that R^{10} does not represent iodine if R^{11} represents hydrogen and with the proviso that R^{10} does not represent trifluoromethyl or difluoromethyl if R^3 and R^{11} represent hydrogen and R^{12} represents methyl.
- R¹⁰ especially preferably represents hydrogen, hydroxyl, formyl, chlorine, methyl, ethyl, methoxy, cyclopropyl, monofluoromethyl, difluoromethyl, dichloromethyl, trifluoromethyl, -CHFCH₃ or difluoromethoxy, with the proviso that R¹⁰ does not represent trifluoromethyl or difluoromethyl if R³ and R¹¹ represent hydrogen and R¹² represents methyl.
- 20 R¹¹ <u>preferably</u> represents hydrogen, chlorine, bromine, iodine, methyl, ethyl, methoxy, ethoxy, methylthio, ethylthio, C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms,
 - R¹¹ particularly preferably represents hydrogen, chlorine, bromine, iodine, methyl or -CHFCH₃.
 - R¹¹ <u>very particularly preferably</u> represents hydrogen, chlorine, methyl or -CHFCH₃.
- 25 R¹² <u>preferably</u> represents hydrogen, methyl, ethyl, n-propyl, isopropyl, C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms, hydroxymethyl, hydroxyethyl, cyclopropyl, cyclopentyl, cyclohexyl or phenyl.
 - R¹² <u>particularly preferably</u> represents hydrogen, methyl, ethyl, isopropyl, trifluoromethyl, difluoromethyl, hydroxymethyl, hydroxyethyl or phenyl.
- 30 R¹² <u>very particularly preferably</u> represents hydrogen, methyl, trifluoromethyl or phenyl.
 - R¹² especially preferably represents methyl.
 - R¹³ and R¹⁴ independently of one another <u>preferably</u> represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

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- R¹³ and R¹⁴ independently of one another <u>particularly preferably</u> represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, difluoromethyl, trifluoromethyl, difluorochloromethyl or trichloromethyl.
- R¹³ and R¹⁴ independently of one another <u>very particularly preferably</u> represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, difluoromethyl, trifluoromethyl or trichloromethyl.
- R¹³ and R¹⁴ especially preferably each represent hydrogen.
- R¹⁵ <u>preferably</u> represents fluorine, chlorine, bromine, iodine, cyano, methyl, ethyl, C₁-C₂-halo-alkyl or C₁-C₂-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.
- 10 R¹⁵ particularly preferably represents fluorine, chlorine, bromine, iodine, cyano, methyl, trifluoromethoxy, difluorochloromethoxy or trichloromethoxy.
 - R¹⁵ <u>very particularly preferably</u> represents fluorine, chlorine, bromine, iodine, methyl, trifluoromethyl or trifluoromethoxy.
 - R¹⁵ especially preferably represents chlorine or methyl.
 - R^{16} and R^{17} independently of one another <u>preferably</u> represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C_1 - C_2 -haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R¹⁶ and R¹⁷ independently of one another <u>particularly preferably</u> represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, difluoromethyl, trifluoromethyl, difluorochloromethyl or trichloromethyl.
 - R¹⁶ and R¹⁷ independently of one another <u>very particularly preferably</u> represent hydrogen, fluorine, chlorine, bromine or methyl.
 - R^{16} and R^{17} especially preferably each represent hydrogen.
- 25 R¹⁸ <u>preferably</u> represents hydrogen, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R¹⁸ particularly preferably represents hydrogen, methyl or trifluoromethyl.
 - R¹⁸ <u>very particularly preferably</u> represents methyl.
- 30 R¹⁹ preferably represents hydrogen, fluorine, chlorine, bromine, iodine, hydroxyl, cyano, C₁-C₄-alkyl, C₁-C₂-haloalkyl, C₁-C₂-haloalkoxy or C₁-C₂-haloalkylthio having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R¹⁹ <u>particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, iodine, hydroxyl, cyano, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, difluoromethyl, trifluoromethyl, trifluoromethyl, difluoromethyl, trifluoromethyl, difluoromethyl, trifluoromethyl, difluoromethyl, trifluoromethyl, difluoromethyl, trifluoromethyl, trifluorometh

- methoxy, difluorochloromethoxy, trichloromethoxy, trifluoromethylthio, difluoromethylthio, difluorochloromethylthio or trichloromethylthio.
- R¹⁹ <u>very particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, iodine, methyl, difluoromethyl, trifluoromethyl or trichloromethyl.
- 5 R¹⁹ especially preferably represents iodine, methyl, difluoromethyl or trifluoromethyl.
 - R²⁰ preferably represents fluorine, chlorine, bromine, iodine, hydroxyl, cyano, C₁-C₄-alkyl, methoxy, ethoxy, methylthio, ethylthio, difluoromethylthio, trifluoromethylthio, C₁-C₂-haloalkyl or C₁-C₂-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.
- 10 R²⁰ particularly preferably represents fluorine, chlorine, bromine, iodine, hydroxyl, cyano, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, trifluoromethyl, difluoromethyl, difluoromethyl, trichloromethyl, methoxy, ethoxy, methylthio, ethylthio, difluoromethylthio, trifluoromethylthio, trifluoromethoxy, difluoromethoxy, difluoromethoxy or trichloromethoxy.
- 15 R²⁰ <u>very particularly preferably</u> represents fluorine, chlorine, bromine, iodine, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.
 - R²¹ preferably represents hydrogen, fluorine, chlorine, bromine, iodine, cyano, C₁-C₄-alkyl, methoxy, ethoxy, methylthio, C₁-C₂-haloalkyl or C₁-C₂-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms, C₁-C₂-alkylsulphinyl or C₁-C₂-alkylsulphonyl.
 - R²¹ particularly preferably represents hydrogen, fluorine, chlorine, bromine, iodine, cyano, n-propyl, isopropyl, n-butyl, iso-butyl, sec-butyl, tert-butyl, trifluoromethyl, difluoromethyl, trichloromethyl, methoxy, ethoxy, methylthio, ethylthio, trifluoromethoxy, difluoromethoxy, difluoromethoxy, trichloromethoxy, methylsulphinyl or methylsulphonyl.
 - R²¹ <u>very particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, iodine, n-propyl, isopropyl, n-butyl, iso-butyl, sec-butyl, tert-butyl, trifluoromethyl, difluoromethyl, trichloromethyl, methylsulphinyl or methylsulphonyl.
 - R²¹ especially preferably represents hydrogen.

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- R²² <u>preferably</u> represents methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
- R²² <u>particularly preferably</u> represents methyl, ethyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
- 35 R²² <u>very particularly preferably</u> represents methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

- R²³ preferably represents methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
- R²³ <u>particularly preferably</u> represents methyl, ethyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
- R²³ <u>very particularly preferably</u> represents methyl, trifluoromethyl, difluoromethyl or trichloromethyl.
- R²⁴ and R²⁵ independently of one another <u>preferably</u> represent hydrogen, fluorine, chlorine, bromine, amino, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R²⁴ and R²⁵ independently of one another <u>particularly preferably</u> represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
- 15 R²⁴ and R²⁵ independently of one another <u>very particularly preferably</u> represent hydrogen, fluorine, chlorine, bromine or methyl.
 - R²⁴ and R²⁵ especially preferably each represent hydrogen.
- R²⁶ <u>preferably</u> represents hydrogen, fluorine, chlorine, bromine, iodine, methyl, ethyl or C₁-C₂20 haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R²⁶ <u>particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, iodine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
 - R²⁶ <u>very particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, methyl or trifluoromethyl.
- R^{26} especially preferably represents methyl or trifluoromethyl.
 - R²⁷ and R²⁸ independently of one another <u>preferably</u> represent hydrogen, fluorine, chlorine, bromine, amino, nitro, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
- 30 R²⁷ and R²⁸ independently of one another <u>particularly preferably</u> represent hydrogen, fluorine, chlorine, bromine, nitro, methyl, ethyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
 - R²⁷ and R²⁸ independently of one another <u>very particularly preferably</u> represent hydrogen, fluorine, chlorine, bromine, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.
 - R²⁷ and R²⁸ especially preferably each represent hydrogen.

- R²⁹ preferably represents fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
- R²⁹ <u>particularly preferably</u> represents fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.
- 5 R²⁹ <u>very particularly preferably</u> represents fluorine, chlorine, bromine, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.
 - R²⁹ especially preferably represents methyl.
- preferably represents hydrogen, fluorine, chlorine, bromine, amino, C₁-C₄-alkylamino, di(C₁-C₄-alkyl)amino, cyano, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R³⁰ <u>particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, cyano, methyl, ethyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
- 15 R³⁰ <u>very particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.
 - R³⁰ especially preferably represents amino, methylamino, dimethylamino, methyl or trifluoromethyl.
- 20 R³¹ <u>preferably</u> represents fluorine, chlorine, bromine, hydroxyl, methyl, ethyl, methoxy, ethoxy, cyclopropyl, C₁-C₂-haloalkyl or C₁-C₂-haloalkoxy having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R³¹ <u>particularly preferably</u> represents fluorine, chlorine, bromine, hydroxyl, methyl, ethyl, methoxy, ethoxy, cyclopropyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
 - R³¹ <u>very particularly preferably</u> represent fluorine, chlorine, bromine, hydroxyl, methyl, methoxy, cyclopropyl, trifluoromethyl, difluoromethyl or trichloromethyl.
- R³² <u>preferably</u> represents hydrogen, fluorine, chlorine, bromine, amino, C₁-C₄-alkylamino, di(C₁-C₄-alkyl)amino, cyano, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R³² <u>particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, cyano, methyl, ethyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
- 35 R³² <u>very particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

- R³² especially preferably represents amino, methylamino, dimethylamino, methyl or trifluoromethyl.
- R³³ preferably represents fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R³³ <u>particularly preferably</u> represents fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
 - R³³ <u>very particularly preferably</u> represents fluorine, chlorine, bromine, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.
- 10 R³³ especially preferably represents methyl, trifluoromethyl or difluoromethyl.
 - R³⁴ preferably represents hydrogen, methyl or ethyl.
 - R³⁴ <u>particularly preferably</u> represents methyl.

- 15 R³⁵ preferably represents fluorine, chlorine, bromine, methyl or ethyl.
 - R³⁵ particularly preferably represents fluorine, chlorine or methyl.
 - R³⁶ preferably represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
- 20 R³⁶ <u>particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, methyl or trifluoromethyl.
 - R³⁷ preferably represents fluorine, chlorine, bromine, iodine, hydroxyl, C₁-C₄-alkyl, methoxy, ethoxy, methylthio, ethylthio, difluoromethylthio, trifluoromethylthio, C₁-C₂-haloalkyl or C₁-C₂-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R³⁷ particularly preferably represents fluorine, chlorine, bromine, iodine, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, trifluoromethyl, difluoromethyl, difluoromethyl, trichloromethyl.
- R³⁷ <u>very particularly preferably</u> represents fluorine, chlorine, bromine, iodine, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.
 - R³⁸ preferably represents hydrogen, methyl, ethyl, C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms, C₁-C₂-alkoxy-C₁-C₂-alkyl, hydroxymethyl, hydroxyethyl, methylsulphonyl or dimethylaminosulphonyl.
- 35 R³⁸ <u>particularly preferably</u> represents hydrogen, methyl, ethyl, trifluoromethyl, methoxymethyl, ethoxymethyl, hydroxymethyl or hydroxyethyl.

- R³⁸ <u>very particularly preferably</u> represents methyl or methoxymethyl.
- R³⁹ <u>preferably</u> represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
- 5 R³⁹ <u>particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl or trichloromethyl.
 - R³⁹ <u>very particularly preferably</u> represents hydrogen or methyl.
- R⁴⁰ <u>preferably</u> represents hydrogen, fluorine, chlorine, bromine, cyano, methyl, ethyl, isopropyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R⁴⁰ <u>particularly preferably</u> represents hydrogen, fluorine, chlorine, bromine, cyano, methyl, ethyl, isopropyl, trifluoromethyl, difluoromethyl, difluoromethyl or trichloromethyl.
 - R⁴⁰ <u>very particularly preferably</u> represents hydrogen, fluorine, methyl or trifluoromethyl.
- 15 R⁴¹ preferably represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.
 - R⁴¹ particularly preferably represents hydrogen, fluorine, chlorine, bromine, iodine, methyl or trifluoromethyl.
 - R⁴¹ <u>very particularly preferably</u> represents hydrogen or trifluoromethyl.

- R⁴² <u>preferably</u> represents methyl, ethyl, n-propyl or isopropyl.
- R⁴² particularly preferably represents methyl or ethyl.

Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the general meanings given above.

Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the preferred meanings given above.

Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the particularly preferred meanings given above.

Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the very particularly preferred meanings given above.

Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the especially preferred meanings given above.

Emphasis is given to compounds of the formula (I) in which L represents L-2.

Emphasis is given to compounds of the formula (I) in which R¹ represents hydrogen. Emphasis is given to compounds of the formula (I) in which R¹ represents formyl.

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Emphasis is furthermore given to compounds of the formula (I) in which R^1 represents $-C(=O)C(=O)R^4$ where R^4 is as defined above.

Emphasis is given to compounds of the formula (I) in which A represents A1.

Emphasis is given to compounds of the formula (I) in which R³ represents hydrogen.

Emphasis is given to compounds of the formula (I) in which R³ represents halogen, preferably fluorine, chlorine, bromine or iodine, particularly preferably fluorine, chlorine or bromine, very particularly preferably fluorine or chlorine.

Emphasis is given to compounds of the formula (I) in which R^3 represents C_1 - C_8 -alkyl, preferably C_1 - C_6 -alkyl, particularly preferably methyl, ethyl, n-, iso-, sec- or tert-butyl, very particularly preferably methyl or ethyl.

Emphasis is given to compounds of the formula (I) in which R^3 represents C_1 - C_8 -haloalkyl, preferably C_1 - C_6 -haloalkyl having 1 to 13 fluorine, chlorine and/or bromine atoms, particularly preferably C_1 - C_4 -haloalkyl having 1 to 9 fluorine, chlorine and/or bromine atoms, very particularly preferably trifluoromethyl.

Saturated or unsaturated hydrocarbon radicals, such as alkyl or alkenyl, can in each case be straightchain or branched, as far as this is possible, including in combination with heteroatoms, such as, for example, in alkoxy.

Optionally substituted radicals can be mono- or polysubstituted, where in the case of polysubstitution the substituents can be identical or different.

Halogen-substituted radicals, such as, for example, haloalkyl, are mono- or polyhalogenated. In the case of polyhalogenation, the halogen atoms can be identical or different. Here, halogen represents fluorine, chlorine, bromine and iodine, in particular fluorine, chlorine and bromine.

However, the general or preferred radical definitions or illustrations given above can also be combined with one another as desired, i.e. between the respective ranges and preferred ranges. The definitions apply both to the end products and, correspondingly, to the precursors and intermediates.

30 The definitions mentioned can be combined with one another as desired. Moreover, individual definitions may not apply.

Preference, particular preference or very particular preference is given to the compounds of the formula (I) which carry the substituents mentioned as being preferred, particularly preferred and very particularly preferred, respectively.

<u>Description of the processes according to the invention for preparing the isopentylcarbox-anilides of the formula (I) and the intermediates</u>

Process (a)

Using 5-fluoro-1,3-dimethyl-1H-pyrazole-4-carbonyl chloride and [2-(3-methylbutyl)phenyl]amine as starting materials, the process (a) according to the invention can be illustrated by the following formula scheme:

The formula (II) provides a general definition of the carboxylic acid derivatives required as starting materials for carrying out the process (a) according to the invention. In this formula (II), A preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for A. X¹ preferably represents chlorine, bromine or hydroxyl.

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The carboxylic acid derivatives of the formula (II) are known and/or can be prepared by known processes (cf. WO 93/11117, EP-A 0 545 099, EP-A 0 589 301 and EP-A 0 589 313).

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The formula (III) provides a general definition of the aniline derivatives furthermore required as starting materials for carrying out the process (a) according to the invention. In this formula (III), L, R¹ and R³ preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for these radicals.

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Some of the aniline derivatives of the formula (III) in which L represents L-1 are novel. Aniline derivatives of the formula (III) in which L represents L-1 can be prepared by

f) reacting cyanoanilines of the formula (VIII)

$$R^2$$
 (VIII)

in which R1 and R2 are as defined above

in a first step with a Grignard reagent of the formula (IX)

$$R^3$$
 (IX)

in which

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R³ is as defined above,

X³ represents chlorine, bromine or iodine,

if appropriate in the presence of a diluent, and reacting the resulting alkanoneanilines of the formula (X)

$$R^{1}$$
 R^{1}
 R^{3}
 CH_{3}
 (X)

in which R¹, R² and R³ are as defined above

in a second step with hydrazine (or hydrazine hydrate) in the presence of a base (for example alkali metal or alkaline earth metal hydroxides, such as sodium hydroxide or potassium hydroxide) and, if appropriate, in the presence of a diluent.

The formula (VIII) provides a general definition of the cyanoanilines required as starting materials for carrying out the process (f) according to the invention. In this formula (VIII), R¹ and R² preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for these radicals.

The cyanoanilines of the formula (VIII) are known and/or can be prepared by known processes. Cyanoanilines of the formula (VIII) in which R¹ does not represent hydrogen can be obtained by reacting cyanoanilines of the formula (V-a)

$$H_2N$$
 R^2 $(V-a)$

in which R² is as defined above with halides of the formula (IV)

$$R^{1-A} = X^2$$
 (IV)

5 in which R^{1-A} is as defined above

in the presence of a base and in the presence of a diluent. (The reaction conditions of the process (b) apply correspondingly.)

The formula (IX) provides a general definition of the Grignard reagents furthermore required as starting materials for carrying out the process (f) according to the invention. In this formula (IX), R³ preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for this radical. X³ preferably represents bromine.

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The Grignard reagents of the formula (IX) are known or can be obtained by known processes.

The alkanoneanilines of the formula (X), which are intermediates in the process (f) according to the invention, are novel and also form part of the subject-matter of this application. In the formula (X), the radicals R¹, R² and R³ preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for these radicals.

Hydrazine (or hydrazine hydrate), which is also required as a reagent for the process (f) according to the invention, is a known chemical for synthesis.

The process (f) according to the invention can be carried out in various variants. Thus, it is possible to initially convert cyanoanilines of the formula (V-a) into the corresponding alkanoneanilines of the formula (VII-a)

$$H_2N$$
 R^3
 CH_3
 CH_3
 $(VII-a)$

in which R² and R³ are as defined above,

which are then, if appropriate, reacted with halides of the formula (IV)

$$R^{1-A} X^2$$
 (IV)

5 in which R^{1-A} is as defined above

in the presence of a base and in the presence of a diluent, to give the corresponding alkanoneanilines of the formula (X). [The reaction conditions of the process (b) apply correspondingly.]

However, it is also possible to convert the alkanoneanilines of the formula (VII-a) according to process (f) according to the invention into the corresponding aniline derivatives of the formula (III-a)

$$H_2N$$
 R^2
 R^3
 CH_3

in which R² and R³ are as defined above,

which are then, if appropriate, reacted with halides of the formula (IV)

$$R^{1-A} X^2$$
 (IV)

in which R1-A is as defined above

in the presence of a base and in the presence of a diluent to give the corresponding aniline derivatives of the formula (III). [The reaction conditions of process (b) apply correspondingly.]

20 Aniline derivatives of the formula (III-b)

$$R^{3-B}$$
 (IIIb)

in which

 R^{1-B} a) represents C₁-C₈-alkyl, C₁-C₆-alkylsulphinyl, C₁-C₆-alkylsulphonyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₁-C₈-cycloalkyl; C₁-C₆-haloalkyl, C₁-C₄-haloalkylsulphinyl, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; for-5 myl, formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl; halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms; (C₁-C₈-alkyl)carbonyl, (C₁-C₈-alkoxy)carbonyl, (C₁-C₄-alkoxy-C₁-C₄-alkyl)carbonyl, (C₃-C₈-cycloalkyl)carbonyl; (C₁-C₆-haloalkyl)carbonyl, (C₁-C₆-haloalkoxy)carbonyl, 10 (halo-C₁-C₄-alkoxy-C₁-C₄-alkyl)carbonyl, (C₃-C₈-halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O)R⁴, CONR⁵R⁶ or -CH₂NR⁷R⁸, and

R^{3-B} represents hydrogen, halogen, C₁-C₈-alkyl, C₁-C₈-haloalkyl,

or

 R^{1-B} 15 b) represents hydrogen, C₁-C₈-alkyl, C₁-C₆-alkylsulphinyl, C₁-C₆-alkylsulphonyl, C₁-C₄alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl; C₁-C₆-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄haloalkylsulphinyl, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbo-20 nyl-C₁-C₃-alkyl; halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms; $(C_1-C_8$ -alkyl)carbonyl, $(C_1-C_8$ -alkoxy)carbonyl, $(C_1-C_4$ -alkoxy- C_1-C_4 -alkyl)carbonyl, $(C_3-C_8$ -cycloalkyl)carbonyl; $(C_1-C_6$ -haloalkyl)carbonyl, $(C_1-C_6$ -haloalkoxy)carbonyl, (halo-C₁-C₄-alkoxy-C₁-C₄-alkyl)carbonyl, (C₃-C₈-halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O)R⁴, 25 CONR⁵R⁶ or -CH₂NR⁷R⁸ steht, and

R^{3-B} represents halogen, C₁-C₈-alkyl, C₁-C₈-haloalkyl,

and

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R², R⁴, R⁵, R⁶, R⁷ and R⁸ are each as defined above

are novel and also form part of the subject-matter of this application.

The preferred, particularly preferred and very particularly preferred meanings of R¹ and R³ apply correspondingly to R^{1-B} and R^{3-B}, where in case a) R^{1-B} does in each case not represent hydrogen and in case b) R^{3-B} does not represent hydrogen. The preferred, particularly preferred and very particularly preferred meanings of R², R⁴, R⁵, R⁶, R⁷ and R⁸ apply likewise to the novel compounds of the formula (III-b).

Emphasis is given to compounds of the formula (III-b) in which R^1 and R^2 each represent hydrogen and R^3 represents fluorine, chlorine, methyl, ethyl, trifluoromethyl and pentafluoroethyl.

Aniline derivatives of the formula (III) in which L represents L-1 are furthermore obtained by

g) reacting aniline halides of the formula (XI)

in which

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R^{1-A} and R² are as defined above and

X⁴ represents halogen

in a first step with alkynes of the formula (XII)

$$H \xrightarrow{\mathbb{R}^3 \setminus \mathbb{CH}_3} (XII)$$

in which R3 is as defined above

in the presence of a catalyst, if appropriate in the presence of a base and if appropriate in the presence of a diluent, and, in a second step, hydrogenating the resulting alkyneanilines of the formula (XIII)

in which R1-A, R2 and R3 are as defined above,

if appropriate in the presence of a diluent and if appropriate in the presence of a catalyst.

The formula (XI) provides a general definition of the aniline halides required as starting materials for carrying out the process (g) according to the invention. In this formula (XI), R² preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred for this radical.

R^{1-A} has preferably, particularly preferably and very particularly preferably those meanings which have been mentioned in connection with the description of the compounds of the formula (IV) as being preferred, particularly preferred and very particularly preferred, respectively, for this radical.

The aniline halides of the formula (XI) are known and/or can be obtained by known processes, for example from the corresponding derivatives which are unsubstituted at the nitrogen by reaction with the halides of the formula (IV).

The formula (XII) provides a general definition of the alkynes furthermore required as starting materials for carrying out the process (g) according to the invention. In this formula (XII), R³ preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for this radical.

The alkynes of the formula (XII) are known.

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obtained when carrying out the process (g) according to the invention. In this formula (XIII), R² and R³ preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for these radicals. R^{1-A} has preferably, particularly preferably and very particularly preferably those meanings which have been mentioned in connection with the description of the compounds of the formula (IV) as being preferred, particularly preferred and very particularly preferred, respectively, for this radical.

Some of the alkyneanilines of the formula (XIII) are known. They are obtained by process (g) according to the invention.

If aniline derivatives of the formula (III) in which R¹ represents hydrogen are to be obtained, R^{1-A} is chosen such that it acts as a protective group which, after process (g) according to the invention, can be removed by customary methods.

The aniline derivatives of the formula (III) in which L represents L-2, L-3 or L-4 are known and/or can be obtained by known processes (cf., for example, EP-A 1 036 793 and EP-A 0 737 682).

Aniline derivatives of the formula (III) in which L represents L-2, L-3 or L-4 and R¹ does not represent hydrogen can be obtained by reacting anilines of the formula (III-c)

$$H_2N$$
 CH_3
 CH_3
 CH_3
 CH_3

in which

 L^1 represents L-2, L-3 or L-4 and

L-2, L-3, L-4 und R³ are as defined above

5 with halides of the formula (IV)

$$R^{1-A} X^2$$
 (IV)

in which R^{1-A} and X² are as defined above

in the presence of a base and in the presence of a diluent. [The reaction conditions of process (b) apply correspondingly.]

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Process (b)

Using 5-fluoro-1,3-dimethyl-N-[2-(3-methylbutyl)phenyl]-1H-pyrazole-4-carboxamide and ethyl chloro(oxo)acetate as starting materials, the course of the process (b) according to the invention can be illustrated by the following formula scheme:

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$$H_3C$$
 CH_3
 H_3C
 CH_3
 H_3C
 CH_3
 H_3C
 C_2H_5
 C_2H_5
 C_2H_5
 C_2H_5

The formula (I-a) provides a general definition of the isopentylcarboxanilides required as starting materials for carrying out the process (b) according to the invention. In this formula (I-a), R², R³ and A preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for these radicals.

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The isopentylcarboxanilides of the formula (I-a) are also compounds according to the invention and also form part of the subject-matter of this application. They can be obtained by one of processes (a). (c), (d) or (e) according to the invention (where $R^1 = hydrogen$).

The formula (IV) provides a general definition of the halides furthermore required as starting materials for carrying out the process (b) according to the invention.

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preferably represents C₁-C₆-alkyl, C₁-C₄-alkylsulphinyl, C₁-C₄-alkylsulphonyl, C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-cycloalkyl; C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl; halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms;

(C_1 - C_6 -alkyl)carbonyl, (C_1 - C_4 -alkoxy)carbonyl, (C_1 - C_3 -alkoxy- C_1 - C_3 -alkyl)carbonyl, (C_3 - C_6 -cycloalkyl)carbonyl; (C_1 - C_4 -haloalkyl)carbonyl, (C_1 - C_4 -haloalkoxy)carbonyl, (halo- C_1 - C_3 -alkyl)carbonyl, (C_3 - C_6 -halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O) R^4 , -CON R^5 R^6 or -CH₂N R^7 R^8 .

- R^{1-A} particularly preferably represents methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl, pentyl or hexyl, methylsulphinyl, ethylsulphinyl, n- or isopropylsulphinyl, n-, iso-, sec- or tert-butylsulphinyl, methylsulphonyl, ethylsulphonyl, n- or isopropylsulphonyl, n-, iso-, sec-15 methoxymethyl, methoxyethyl, ethoxymethyl, or tert-butylsulphonyl, cyclopropyl, cyclopentyl, cyclohexyl, trifluoromethyl, trichloromethyl, trifluoroethyl, difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethylsulphinyl, trifluoromethylsulphonyl, trifluoromethoxymethyl; formyl, -CH2-CHO, -(CH2)2-CHO, -CH₂-CO-CH₃, -CH₂-CO-CH₂CH₃, -CH₂-CO-CH(CH₃)₂, -(CH₂)₂-CO-CH₃, 20 -(CH₂)₂-CO-CH₂CH₃, -(CH₂)₂-CO-CH(CH₃)₂, -CH₂-CO₂CH₃, -CH₂-CO₂CH₂CH₃, -CH₂-CO₂CH(CH₃)₂, -(CH₂)₂-CO₂CH₃, -(CH₂)₂-CO₂CH₂CH₃, -(CH₂)₂-CO₂CH(CH₃)₂, -CH₂-CO-CF₃, -CH₂-CO-CCl₃, -CH₂-CO-CH₂CF₃, -CH₂-CO-CH₂CCl₃, -(CH₂)₂-CO-CH₂CF₃, -(CH₂)₂-CO-CH₂CCl₃, -CH₂-CO₂CH₂CF₃, -CH₂-CO₂CF₂CF₃, -CH₂-CO₂CH₂CCl₃, -CH₂-CO₂CCl₂CCl₃, -(CH₂)₂-CO₂CH₂CF₃, <math>-(CH₂)₂-CO₂CF₂CF₃-(CH₂)₂-CO₂CH₂CCl₃, 25 -(CH₂)₂-CO₂CCl₂CCl₃;
 - methylcarbonyl, ethylcarbonyl, n-propylcarbonyl, isopropylcarbonyl, tert-butylcarbonyl, methoxycarbonyl, ethoxycarbonyl, tert-butoxycarbonyl, cyclopropylcarbonyl; trifluoromethylcarbonyl, trifluoromethoxycarbonyl, or $-C(=O)C(=O)R^5$, $-CONR^6R^7$ or $-CH_2NR^8R^9$.
- 30 R^{1-A} <u>very particularly preferably</u> represents methyl, methoxymethyl, formyl, -CH₂-CHO, -(CH₂)₂-CHO, -CH₂-CO-CH₃, -CH₂-CO-CH₂CH₃, -CH₂-CO-CH(CH₃)₂, -C(=O)CHO, -C(=O)C(=O)CH₂OCH₃, -C(=O)CO₂CH₃, -C(=O)CO₂CH₂CH₃.
 - X² <u>preferably</u> represents chlorine or bromine.
- 35 Halides of the formula (IV) are known.

Process (c)

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Using 2-iodo-N-[2-(3-methylbutanoyl)phenyl]benzamide as starting material, and hydrazine and a base, the course of the process (c) according to the invention can be illustrated by the formula scheme below:

The formula (V) provides a general definition of the isopentone derivatives required as starting materials for carrying out the process (c) according to the invention. In this formula (V), R¹, R², R³ and A preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for these radicals.

The isopentone derivatives of the formula (V) are novel. They are obtained by

h) reacting carboxylic acid derivatives of the formula (II)

$$A \xrightarrow{O} X^1 \qquad (II),$$

in which

A is as defined above

X¹ represents halogen or hydroxyl with alkanoneanilines of the formula (X)

$$R^{1}$$
 R^{1}
 R^{3}
 R^{3}
 CH_{3}
 CH_{3}
 CH_{3}

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in which R¹, R² and R³ are as defined above,

if appropriate in the presence of a catalyst, if appropriate in the presence of a condensing agent, if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent.

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The carboxylic acid derivatives of the formula (II) required as starting materials for carrying out the process (h) according to the invention have already been described in connection with process (a) according to the invention.

The alkanoneanilines of the formula (X) furthermore required as starting material for carrying out the process (h) according to the invention have already been described in connection with process (f) according to the invention.

Process (d)

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Using N-{2-[3,3-dimethylbut-1-en-1-yl]phenyl}-5-fluoro-1,3-dimethyl-1H-pyrazole-4-carboxamide as starting material and hydrogen, the course of the process (d) according to the invention can be illustrated by the formula scheme below:

The formula (VI) provides a general definition of the isopentene derivatives required as starting materials for carrying out the process (d) according to the invention. In this formula (VI), R¹, R², R³ and A preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred for these radicals.

The isopentene derivatives of the formula (VI) are novel. They are obtained by

j) reacting carboxamides of the formula (XIV)

in which

R¹, R² and A are as defined above and

X⁵ represents chlorine, bromine, iodine or -OSO₂CF₃ with alkenes of the formula (XV)

$$R^3$$
 CH_3 CH_3 CH_3

25 in which R³ is as defined above

in the presence of a catalyst, if appropriate in the presence of a base and if appropriate in the presence of a diluent.

The formula (XIV) provides a general definition of the carboxamides furthermore required as starting materials for carrying out the process (j) according to the invention. In this formula (XIV), R¹, R² and A preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for these radicals. X⁵ preferably represents bromine or -OSO₂CF₃.

The carboxamides of the formula (XIV) are novel or can be obtained by known methods (cf. WO 02/08195 and WO 02/08197).

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The formula (XV) provides a general definition of the alkenes furthermore required as starting materials for carrying out the process (j) according to the invention. In this formula (XV), R³ preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for this radical.

The alkenes of the formula (XV) are known.

20 Process (e)

Using N-[2-(3,3-dimethylbut-1-yn-1-yl)phenyl]-5-fluoro-1,3-dimethyl-1H-pyrazole-4-carboxamide as starting material and hydrogen, the course of the process (e) according to the invention can be illustrated by the formula scheme below:

$$H_3C$$
 H_3C
 H_3C

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The formula (VII) provides a general definition of the isopentyne derivatives required as starting materials for carrying out the process (e) according to the invention. In this formula (VII), R¹, R², R³ and A preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred for these radicals.

The isopentyne derivatives of the formula (VII) in which A does not represent A1 are novel. The isopentyne derivatives of the formula (VII) are obtained by

k) reacting carboxamides of the formula (XIV)

5 in which

R¹, R² and A are as defined above and

X⁵ represents chlorine, bromine, iodine or -OSO₂CF₃, with alkynes of the formula (XII)

$$H \xrightarrow{R_3} CH_3$$
 (XII)

in which R³ is as defined above

in the presence of a catalyst, if appropriate in the presence of a base and if appropriate in the presence of a diluent.

The carboxamides of the formula (XIV) required as starting materials for carrying out the process (k) according to the invention have already been described in connection with the process (j) according to the invention.

The alkynes of the formula (XII) furthermore required as starting materials for carrying out the process (k) according to the invention have already been described in connection with the process (g) according to the invention.

Reaction conditions

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Suitable diluents for carrying out the process (a) and (h) according to the invention are all inert organic solvents. These preferably include aliphatic, alicyclic or aromatic hydrocarbons, such as, for example, petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane, benzene, toluene, xylene or decalin; halogenated hydrocarbons, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; ethers, such as diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole, or amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide.

The processes (a) and (h) according to the invention are, if appropriate, carried out in the presence of a suitable acid acceptor. Suitable acid acceptors are all customary inorganic or organic bases. These preferably include alkaline earth metal or alkali metal hydrides, hydroxides, amides, alkoxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydroxide, sodium amide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate, sodium bicarbonate or ammonium carbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylamiline, N,N-dimethylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

The processes (a) and (h) according to the invention are, if appropriate, carried out in the presence of a suitable condensing agent. Suitable condensing agents are all condensing agents customarily used for such amidation reactions. Acid halide formers, such as phosgene, phosphorus tribromide, phosphorus trichloride, phosphorus pentachloride, phosphorus oxychloride or thionyl chloride; anhydride formers, such as ethyl chloroformate, methyl chloroformate, isopropyl chloroformate, isobutyl chloroformate or methylsulphonyl chloride; carbodiimides, such as N,N'-dicyclohexylcarbodiimide (DCC), or other customary condensing agents, such as phosphorus pentoxide, polyphosphoric acid, N,N'-carbonyldiimidazole, 2-ethoxy-N-ethoxycarbonyl-1,2-dihydroquinoline (EEDQ), triphenylphosphine/carbon tetrachloride or bromotripyrrolidinophosphonium hexafluorophosphate may be mentioned by way of example.

The processes (a) and (h) according to the invention are, if appropriate, carried out in the presence of a catalyst. Examples which may be mentioned are 4-dimethylaminopyridine, 1-hydroxybenzotriazole or dimethylformamide.

When carrying out the processes (a) and (h) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the processes are carried out at temperatures of from 0°C to 150°C, preferably at temperatures of from 0°C to 80°C.

For carrying out the process (a) according to the invention for preparing the compounds of the formula (I), in general from 0.2 to 5 mol, preferably from 0.5 to 2 mol, of aniline derivative of the formula (III) are employed per mole of the carboxylic acid derivative of the formula (II).

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For carrying out the process (h) according to the invention for preparing the compounds of the formula (V), in general from 0.2 to 5 mol, preferably from 0.5 to 2 mol, of alkanoneaniline of the formula (X) are employed per mole of the carboxylic acid derivative of the formula (II).

Suitable diluents for carrying out the process (b) according to the invention are all inert organic solvents. These preferably include aliphatic, alicyclic or aromatic hydrocarbons, such as, for example, petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane, benzene, toluene, xylene or decalin; halogenated hydrocarbons, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; ethers, such as diethyl ether, diisopropyl ether, methyl tert-butyl ether, methyl tert-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole, or amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide.

The process (b) according to the invention is carried out in the presence of a base. Suitable bases are all customary inorganic or organic bases. These preferably include alkaline earth metal or alkali metal hydrides, hydroxides, amides, alkoxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydride, sodium amide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate, sodium bicarbonate or caesium carbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylamiline, N,N-dimethylbenzylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

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When carrying out the process (b) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the process is carried out at temperatures of from 0°C to 150°C, preferably at temperatures of from 20°C to 110°C.

For carrying out the process (b) according to the invention for preparing the compounds of the formula (I), in general from 0.2 to 5 mol, preferably from 0.5 to 2 mol, of halide of the formula (IV) are employed per mole of the isopentylcarboxanilide of the formula (I-a).

Suitable diluents for carrying out the process (c) according to the invention and the second step of process (f) are all inert organic solvents. These preferably include aliphatic, alicyclic or aromatic hydrocarbons, such as, for example, petroleum ether, hexane, heptane, cyclohexane, methylcyclo-

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hexane, benzene, toluene, xylene or decalin; halogenated hydrocarbons, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; ethers, such as diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole; ketones, such as acetone, butanone, methyl isobutyl ketone or cyclohexanone; nitriles, such as acetonitrile, propionitrile, n- or i-butyronitrile or benzonitrile; amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide; sulphoxides, such as dimethyl sulphoxide; sulphones, such as sulpholane; alcohols, such as methanol, ethanol, n- or i-propanol, n-, i-, sec- or tert-butanol, ethanediol, propane-1,2-diol, ethoxyethanol, methoxyethanol, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, triethylene glycol, mixtures thereof with water or pure water.

The process (c) according to the invention and the second step of process (f) are carried out in the presence of a base. Preferred bases are alkaline earth metal or alkali metal hydroxides, such as, for example, sodium hydroxide, potassium hydroxide, ammonium hydroxide.

When carrying out the process (c) according to the invention and the second step of process (f), the reaction temperatures can be varied within a relatively wide range. In general, the reactions are carried out at temperatures of from 100°C to 300°C, preferably at temperatures of from 150°C to 250°C.

When carrying out the process (c) according to the invention for preparing the compounds of the formula (I), in general from 0.2 to 5 mol, preferably from 0.5 to 3 mol, of hydrazine (or hydrazine

hydrate) are employed per mole of the isopentone derivative of the formula (V).

For carrying out the second step of process (f) for preparing the compounds of the aniline derivatives of the formula (III), in general from 0.2 to 5 mol, preferably from 0.5 to 3 mol, of hydrazine (or hydrazine hydrate) are employed per mole of the alkanoneaniline of the formula (X).

Suitable diluents for carrying out the processes (d) and (e) according to the invention and the second step of process (g) are all inert organic solvents. These preferably include aliphatic or alicyclic hydrocarbons, such as, for example, petroleum ether, hexane, heptane, cyclohexane, methyl-cyclohexane or decalin; ethers, such as diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane or 1,2-diethoxyethane; alcohols, such as methanol, ethanol, n- or isopropanol, n-, iso-, sec- or tert-butanol, ethanediol, propane-1,2-diol, ethoxyethanol, methoxyethanol, diethylene glycol monomethyl ether, diethylene glycol monomethyl ether, mixtures thereof with water or pure water.

The processes (d) and (e) according to the invention and the second step of process (g) are carried out in the presence of a catalyst. Suitable catalysts are all catalysts which are usually used for hydrogenation. The following may be mentioned by way of example: Raney nickel, palladium or platinum, if appropriate on a support, such as, for example, activated carbon.

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Instead of in the presence of hydrogen in combination with a catalyst, the hydrogenation in the processes (d) and (e) according to the invention and in the second step of process (g) can also be carried out in the presence of triethylsilane.

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When carrying out the processes (d) and (e) according to the invention and the second step of process (g), the reaction temperatures can be varied within a relatively wide range. In general, the reactions are carried out at temperatures of from 0°C to 150°C, preferably at temperatures of from 20°C to 100°C.

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The processes (d) and (e) according to the invention and the second step of process (g) are carried out under a hydrogen pressure between 0.5 and 200 bar, preferably between 2 and 50 bar, particularly preferably between 3 and 10 bar.

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Suitable diluents for carrying out the first step of process (f) are all inert organic solvents. These preferably include aliphatic or alicyclic hydrocarbons, such as, for example, petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane or decalin; ethers, such as diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane or 1,2-diethoxyethane.

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When carrying out the first step of process (f), the reaction temperatures can be varied within a relatively wide range. In general, the reaction is carried out at temperatures from 0°C to 200°C, preferably at temperatures of from 20°C to 150°C.

For carrying out the first step of process (f) for preparing the compounds of the alkanoneanilines of the formula (X), in general from 0.2 to 5 mol, preferably from 0.5 to 3 mol, of Grignard reagent of the formula (IX) are employed per mole of the cyanoaniline of the formula (VIII).

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Suitable diluents for carrying out the first step of process (g) and the processes (j) and (k) according to the invention are all inert organic solvents. These preferably include nitriles, such as acetonitrile, propionitrile, n- or i-butyronitrile or benzonitrile, or amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric

triamide; ethers, such as diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane or 1,2-diethoxyethane.

The first step of process (g) and the processes (j) and (k) according to the invention are, if appropriate, carried out in the presence of a suitable acid acceptor. Suitable acid acceptors are all customary inorganic or organic bases. These preferably include alkaline earth metal or alkali metal hydrides, hydroxides, amides, alkoxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydride, sodium amide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate, sodium bicarbonate or ammonium carbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylamiline, N,N-dimethylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

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The first step of process (g) and the processes (j) and (k) according to the invention are carried out in the presence of one or more catalysts.

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Suitable catalysts are in particular palladium salts or complexes. Suitable for this purpose are, preferably, palladium chloride, palladium acetate, tetrakis(triphenylphosphine)palladium or bis(triphenylphosphine)palladium dichloride. It is also possible to generate a palladium complex in the reaction mixture by adding a palladium salt and a complex ligand separately to the reaction.

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Suitable ligands are, preferably, organophosphorus compounds. The following may be mentioned by way of example: triphenylphosphine, tri-o-tolylphosphine, 2,2'-bis(diphenylphosphino)-1,1'-binaphthyl, dicyclohexylphosphinebiphenyl, 1,4-bis(diphenylphosphino)butane, bisdiphenylphosphinoferrocene, di-(tert-butylphosphino)biphenyl, di(cyclohexylphosphino)biphenyl, 2-dicyclohexylphosphino-2'-N,N-dimethylaminobiphenyl, tricyclohexylphosphine, tri-tert.-butylphosphine. However, ligands may also be dispensed with.

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The first step of process (g) and the processes (j) and (k) according to the invention are furthermore, if appropriate, carried out in the presence of a further metal salt, such as copper salts, for example copper(I) iodide.

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When carrying out the first step of process (g) and the processes (j) and (k) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the

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reactions are carried out at temperatures of from 20°C to 180°C, preferably at temperatures of from 50°C to 150°C.

For carrying out the first step of process (g) for preparing the aniline derivatives of the formula (III), in general from 1 to 5 mol, preferably from 1 to 3 mol, of alkyne of the formula (XII) are employed per mole of the aniline halide of the formula (XI).

For carrying out the process (j) according to the invention for preparing the isopentene derivatives of the formula (VI), in general from 1 to 5 mol, preferably from 1 to 3 mol, of alkene of the formula (XV) are employed per mole of the carboxamide of the formula (XIV).

For carrying out the process (k) according to the invention for preparing the isopentyne derivatives of the formula (VII), in general from 1 to 5 mol, preferably from 1 to 3 mol, of alkyne of the formula (XII) are employed per mole of the carboxamide of the formula (XIV).

Unless stated otherwise, all processes according to the invention are generally carried out under atmospheric pressure. However, it is also possible to operate under elevated or reduced pressure - in general between 0.1 bar and 10 bar.

The substances according to the invention have potent microbial activity and can be employed for controlling unwanted microorganisms, such as fungi and bacteria, in crop protection and in the protection of materials.

Fungicides can be employed in crop protection for controlling Plasmodiophoromycetes, Oomycetes, Chytridiomycetes, Zygomycetes, Ascomycetes, Basidiomycetes and Deuteromycetes.

Bactericides can be employed in crop protection for controlling Pseudomonadaceae, Rhizobiaceae, Enterobacteriaceae, Corynebacteriaceae and Streptomycetaceae.

Some pathogens causing fungal and bacterial diseases which come under the generic names listed above may be mentioned as examples, but not by way of limitation:

Xanthomonas species, such as, for example, Xanthomonas campestris pv. oryzae;

Pseudomonas species, such as, for example, Pseudomonas syringae pv. lachrymans;

Erwinia species, such as, for example, Erwinia amylovora;

Pythium species, such as, for example, Pythium ultimum;Phytophthora species, such as, for example, Phytophthora infestans;

Pseudoperonospora species, such as, for example, Pseudoperonospora humuli or Pseudoperonospora cubensis;

Plasmopara species, such as, for example, Plasmopara viticola;

Bremia species, such as, for example, Bremia lactucae;

5 Peronospora species, such as, for example, Peronospora pisi or P. brassicae;

Erysiphe species, such as, for example, Erysiphe graminis;

Sphaerotheca species, such as, for example, Sphaerotheca fuliginea;

Podosphaera species, such as, for example, Podosphaera leucotricha;

Venturia species, such as, for example, Venturia inaequalis;

10 Pyrenophora species, such as, for example, Pyrenophora teres or P. graminea

(conidia form: Drechslera, syn: Helminthosporium);

Cochliobolus species, such as, for example, Cochliobolus sativus

(conidia form: Drechslera, syn: Helminthosporium);

Uromyces species, such as, for example, Uromyces appendiculatus;

15 Puccinia species, such as, for example, Puccinia recondita;

Sclerotinia species, such as, for example, Sclerotinia sclerotiorum:

Tilletia species, such as, for example, Tilletia caries;

Ustilago species, such as, for example, Ustilago nuda or Ustilago avenae;

Pellicularia species, such as, for example, Pellicularia sasakii:

20 Pyricularia species, such as, for example, Pyricularia oryzae;

Fusarium species, such as, for example, Fusarium culmorum;

Botrytis species, such as, for example, Botrytis cinerea;

Septoria species, such as, for example, Septoria nodorum;

Leptosphaeria species, such as, for example, Leptosphaeria nodorum;

25 Cercospora species, such as, for example, Cercospora canescens;

Alternaria species, such as, for example, Alternaria brassicae; and

Pseudocercosporella species, such as, for example, Pseudocercosporella herpotrichoides,

Rhizoctonia species, such as, for example, Rhizoctonia solani.

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The active compounds according to the invention also show a strong invigorating action in plants. Accordingly, they are suitable for mobilizing the internal defences of the plant against attack by unwanted microorganisms.

In the present context, plant-invigorating (resistance-inducing) compounds are to be understood as meaning substances which are capable of stimulating the defence system of plants such that, when the 5

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treated plants are subsequently inoculated with unwanted microorganisms, they display substantial resistance to these microorganisms.

In the present case, unwanted microorganisms are to be understood as meaning phytopathogenic fungi, bacteria and viruses. The compounds according to the invention can thus be used to protect plants within a certain period of time after treatment against attack by the pathogens mentioned. The period of time for which this protection is achieved generally extends for 1 to 10 days, preferably 1 to 7 days, from the treatment of the plants with the active compounds.

The fact that the active compounds are well tolerated by plants at the concentrations required for controlling plant diseases permits the treatment of above-ground parts of plants, of propagation stock and seeds, and of the soil.

Here, the active compounds according to the invention can be used with particularly good results for controlling cereal diseases, such as, for example, against Puccinia species, and of diseases in viticulture and in the cultivation of fruits and vegetables, such as, for example, against botrytis, Venturia or Alternaria species.

The active compounds according to the invention are also suitable for increasing the yield of crops. In addition, they show reduced toxicity and are well tolerated by plants.

If appropriate, the active compounds according to the invention can, at certain concentrations and application rates, also be employed as herbicides, for regulating plant growth and for controlling animal pests. If appropriate, they can also be used as intermediates or precursors in the synthesis of other active compounds.

According to the invention, it is possible to treat all plants and parts of plants. Plants are to be understood here as meaning all plants and plant populations, such as desired and undesired wild plants or crop plants (including naturally occurring crop plants). Crop plants can be plants which can be obtained by conventional breeding and optimization methods or by biotechnological and genetic engineering methods or combinations of these methods, including the transgenic plants and including plant cultivars which can or cannot be protected by plant breeders' certificates. Parts of plants are to be understood as meaning all above-ground and below-ground parts and organs of plants, such as shoot, leaf, flower and root, examples which may be mentioned being leaves, needles, stems, trunks, flowers, fruit-bodies, fruits and seeds and also roots, tubers and rhizomes. Parts of plants also include harvested material and vegetative and generative propagation material, for example seedlings, tubers, rhizomes, cuttings and seeds.

The treatment of the plants and parts of plants according to the invention with the active compounds is carried out directly or by action on their environment, habitat or storage area according to customary treatment methods, for example by dipping, spraying, evaporating, atomizing, broadcasting, brushing-on and, in the case of propagation material, in particular in the case of seeds, furthermore by one- or multilayer coating.

In the protection of materials, the compounds according to the invention can be employed for protecting industrial materials against infection with, and destruction by, unwanted microorganisms.

Industrial materials in the present context are understood as meaning non-living materials which have been prepared for use in industry. For example, industrial materials which are intended to be protected by active compounds according to the invention from microbial change or destruction can be tackifiers, sizes, paper and board, textiles, leather, wood, paints and plastic articles, cooling lubricants and other materials which can be infected with, or destroyed by, microorganisms. Parts of production plants, for example cooling-water circuits, which may be impaired by the proliferation of microorganisms may also be mentioned within the scope of the materials to be protected. Industrial materials which may be mentioned within the scope of the present invention are preferably tackifiers, sizes, paper and board, leather, wood, paints, cooling lubricants and heat-transfer liquids, particularly preferably wood.

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Microorganisms capable of degrading or changing the industrial materials which may be mentioned are, for example, bacteria, fungi, yeasts, algae and slime organisms. The active compounds according to the invention preferably act against fungi, in particular moulds, wood-discolouring and wood-destroying fungi (Basidiomycetes) and against slime organisms and algae.

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Microorganisms of the following genera may be mentioned as examples:

Alternaria, such as Alternaria tenuis,

Aspergillus, such as Aspergillus niger,

Chaetomium, such as Chaetomium globosum,

30 Coniophora, such as Coniophora puetana,

Lentinus, such as Lentinus tigrinus,

Penicillium, such as Penicillium glaucum,

Polyporus, such as Polyporus versicolor.

Aureobasidium, such as Aureobasidium pullulans,

35 Sclerophoma, such as Sclerophoma pityophila,

Trichoderma, such as Trichoderma viride,

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Escherichia, such as Escherichia coli,
Pseudomonas, such as Pseudomonas aeruginosa, and
Staphylococcus, such as Staphylococcus aureus.

- Depending on their particular physical and/or chemical properties, the active compounds can be converted into the customary formulations, such as solutions, emulsions, suspensions, powders, foams, pastes, granules, aerosols and microencapsulations in polymeric substances and in coating compositions for seeds, and ULV cool and warm fogging formulations.
- These formulations are produced in a known manner, for example by mixing the active compounds 10 with extenders, that is liquid solvents, liquefied gases under pressure, and/or solid carriers, optionally with the use of surfactants, that is emulsifiers and/or dispersants, and/or foam formers. If the extender used is water, it is also possible to employ, for example, organic solvents as auxiliary solvents. Essentially, suitable liquid solvents are: aromatics such as xylene, toluene or alkylnaphthalenes, chlorinated aromatics or chlorinated aliphatic hydrocarbons such as chlorobenzenes, chloroethylenes 15 or methylene chloride, aliphatic hydrocarbons such as cyclohexane or paraffins, for example petroleum fractions, alcohols such as butanol or glycol and their ethers and esters, ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents such as dimethylformamide or dimethyl sulphoxide, or else water. Liquefied gaseous extenders or carriers are to be understood as meaning liquids which are gaseous at standard temperature and under 20 atmospheric pressure, for example aerosol propellants such as halogenated hydrocarbons, or else butane, propane, nitrogen and carbon dioxide. Suitable solid carriers are: for example ground natural minerals such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals such as finely divided silica, alumina and silicates. Suitable solid carriers for granules are: for example crushed and fractionated natural rocks such as calcite, marble, 25 pumice, sepiolite and dolomite, or else synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks. Suitable emulsifiers and/or foam formers are: for example nonionic and anionic emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol 30 ethers, alkylsulphonates, alkyl sulphates, arylsulphonates, or else protein hydrolysates. Suitable dispersants are: for example lignosulphite waste liquors and methylcellulose.

Tackifiers such as carboxymethylcellulose, natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, or else natural phospholipids such as cephalins and lecithins and synthetic phospholipids can be used in the formulations. Other possible additives are mineral and vegetable oils.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

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The formulations generally comprise between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

The active compounds according to the invention can, as such or in their formulations, also be used in a mixture with known fungicides, bactericides, acaricides, nematicides or insecticides, to broaden, for example, the activity spectrum or to prevent development of resistance. In many cases, synergistic effects are obtained, i.e. the activity of the mixture is greater than the activity of the individual components.

15 Suitable mixing components are, for example, the following compounds:

Fungicides:

2-phenylphenol; 8-hydroxyquinoline sulphate; acibenzolar-S-methyl; aldimorph; amidoflumet; ampropylfos; ampropylfos-potassium; andoprim; anilazine; azaconazole; azoxystrobin; benalaxyl; benodanil; benomyl; benthiavalicarb-isopropyl; benzamacril; benzamacril-isobutyl; bilanafos; binapacryl; biphenyl; bitertanol; blasticidin-S; bromuconazole; bupirimate; buthiobate; butylamine; calcium polysulphide; capsimycin; captafol; captan; carbendazim; carboxin; carpropamid; carvone; chinomethionat; chlobenthiazone; chlorfenazole; chloroneb; chlorothalonil; chlozolinate; clozylacon; cyazofamid; cyflufenamid; cymoxanil; cyproconazole; cyprodinil; cyprofuram; Dagger G; debacarb; dichlofluanid; dichlone; dichlorophen; diclocymet; diclomezine; dicloran; diethofencarb; difenoconazole; diflumetorim; dimethirimol; dimethomorph; dimoxystrobin; diniconazole; diniconazole-M; dinocap; diphenylamine; dipyrithione; ditalimfos; dithianon; dodine; drazoxolon; edifenphos; epoxiconazole; ethaboxam; ethirimol; etridiazole; famoxadone; fenamidone; fenapanil; fenarimol; fenbuconazole; fenfuram; fenhexamid; fenitropan; fenoxanil; fenpiclonil; fenpropidin; fenpropimorph; ferbam; fluazinam; flubenzimine; fludioxonil; flumetover; flumorph; fluoromide; fluoxastrobin; fluquinconazole; flurprimidol; flusilazole; flusulphamide; flutolanil; flutriafol; folpet; fosetyl-Al; fosetyl-sodium; fuberidazole; fluralaxyl; furametpyr; furcarbanil; furmecyclox; guazatine; hexachlorobenzene; hexaconazole; hymexazole; imazalil; imibenconazole; iminoctadine triacetate; iminoctadine tris(albesil); iodocarb; ipconazole; iprobenfos; iprodione; iprovalicarb; irumamycin; isoprothiolane; isovaledione; kasugamycin; kresoximmethyl; mancozeb; maneb; meferimzone; mepanipyrim; mepronil; metalaxyl; metalaxyl-M; metconazole; methasulphocarb; methfuroxam; metiram; metominostrobin; metsulphoyax; mildiomycin; myclobutanil; myclozolin; natamycin; nicobifen; nitrothal-isopropyl; noviflumuron; nuarimol; ofurace;

orysastrobin; oxadixyl; oxolinic acid; oxpoconazole; oxycarboxin; oxyfenthiin; paclobutrazole; pefurazoate; penconazole; pencycuron; phosdiphen; phthalide; picoxystrobin; piperalin; polyoxins; polyoxorim; probenazole; prochloraz; procymidone; propamocarb; propanosine-sodium; propiconazole; propineb; proquinazid; prothioconazole; pyraclostrobin; pyrazophos; pyrifenox; pyrimethanil; pyroqui-5 lon; pyroxyfur; pyrrolenitrine; quinconazole; quinoxyfen; quintozene; simeconazole; spiroxamine; sulphur; tebuconazole; tecloftalam; tecnazene; tetcyclacis; tetraconazole; thiabendazole; thicyofen; thifluzamide; thiophanate-methyl; thiram; tioxymid; tolclofos-methyl; tolylfluanid; triadimefon; triadimenol; triazbutil; triazoxide; tricyclamide; tricyclazole; tridemorph; trifloxystrobin; triflumizole; triforine; triticonazole; uniconazole; validamycin A; vinclozolin; zineb; ziram; zoxamide; (2S)-N-[2-[4-10 [[3-(4-chlorophenyl)-2-propynyl]oxyl-3-methoxyphenyl]ethyll-3-methyl-2-[(methylsulphonyl)aminolbutanamide; 1-(1-naphthalenyl)-1H-pyrrole-2,5-dione; 2,3,5,6-tetrachloro-4-(methylsulphonyl)pyridine; 2-amino-4-methyl-N-phenyl-5-thiazolecarboxamide; 2-chloro-N-(2,3-dihydro-1,1,3-trimethyl-1H-inden-4-yl)-3-pyridinecarboxamide; 3,4,5-trichloro-2,6-pyridinedicarbonitrile; actinovate; cis-1-(4-chlorophenyl)-2-(1H-1,2,4-triazol-1-yl)cycloheptanol; methyl 1-(2,3-dihydro-2,2-dimethyl-1H-inden-1-yl)-1H-15 imidazole-5-carboxylate; monopotassium carbonate; N-(6-methoxy-3-pyridinyl)-cyclopropanecarboxamide; N-butyl-8-(1,1-dimethylethyl)-1-oxaspiro[4.5]decane-3-amine; sodium tetrathiocarbonate; and copper salts and preparations, such as Bordeaux mixture; copper hydroxide; copper naphthenate; copper oxychloride; copper sulphate; cufraneb; copper oxide; mancopper; oxine-copper.

20 Bactericides:

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bronopol, dichlorophen, nitrapyrin, nickel dimethyldithiocarbamate, kasugamycin, octhilinone, furancarboxylic acid, oxytetracyclin, probenazole, streptomycin, tecloftalam, copper sulphate and other copper preparations.

25 Insecticides / acaricides / nematicides:

abamectin, ABG-9008, acephate, acequinocyl, acetamiprid, acetoprole, acrinathrin, AKD-1022, AKD-3059, AKD-3088, alanycarb, aldicarb, aldoxycarb, allethrin, allethrin 1R-isomers, alpha-cypermethrin (alphamethrin), amidoflumet, aminocarb, amitraz, avermectin, AZ-60541, azadirachtin, azamethiphos, azinphos-methyl, azinphos-ethyl, azocyclotin, Bacillus popilliae, Bacillus sphaericus, Bacillus subtilis, Bacillus thuringiensis, Bacillus thuringiensis strain EG-2348, Bacillus thuringiensis strain GC-91, Bacillus thuringiensis strain NCTC-11821, baculoviruses, Beauveria bassiana, Beauveria tenella, benclothiaz, bendiocarb, benfuracarb, bensultap, benzoximate, beta-cyfluthrin, beta-cypermethrin, bifenazate, bifenthrin, binapacryl, bioallethrin, bioallethrin-S-cyclopentyl-isomer, bioethanomethrin, biopermethrin, bioresmethrin, bistrifluron, BPMC, brofenprox, bromophos-ethyl, bromopropylate, bromfenvinfos (-methyl), BTG-504, BTG-505, bufencarb, buprofezin, butathiofos, butocarboxim, butoxycarboxim, butylpyridaben, cadusafos, camphechlor, carbaryl, carbofuran, carbophenothion,

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carbosulphan, cartap, CGA-50439, chinomethionat, chlordane, chlordimeform, chloethocarb, chlorethoxyfos, chlorfenapyr, chlorfenvinphos, chlorfluazuron, chlormephos, chlorobenzilate, chloropicrin, chlorproxyfen, chlorpyrifos-methyl, chlorpyrifos (-ethyl), chlovaporthrin, chromafenozide, ciscypermethrin, cis-resmethrin, cis-permethrin, clocythrin, cloethocarb, clofentezine, clothianidin, clothiazoben, codlemone, coumaphos, cyanofenphos, cyanophos, cycloprene, cycloprothrin, Cydia pomonella, cyfluthrin, cyhalothrin, cyhexatin, cypermethrin, cyphenothrin (1R-trans-isomer), cyromazine, DDT, deltamethrin, demeton-S-methyl, demeton-S-methylsulphone, diafenthiuron, dialifos, diazinon, dichlofenthion, dichlorvos, dicofol, dicrotophos, dicyclanil, diflubenzuron, dimefluthrin, dimethoate, dimethylvinphos, dinobuton, dinocap, dinotefuran, diofenolan, disulphoton, docusat-sodium, dofenapyn, DOWCO-439, eflusilanate, emamectin, emamectin-benzoate, empenthrin (1R-isomer), endosulphan, Entomopthora spp., EPN, esfenvalerate, ethiofencarb, ethiprole, ethion, ethoprophos, etofenprox, etoxazole, etrimfos, famphur, fenamiphos, fenazaquin, fenbutatin oxide, fenfluthrin, fenitrothion, fenobucarb, fenothiocarb, fenoxacrim, fenoxycarb, fenpropathrin, fenpyrad, fenpyrithrin, fenpyroximate, fensulphothion, fenthion, fentrifanil, fenvalerate, fipronil, flonicamid, fluacrypyrim, fluazuron, flubenzimine, flubrocythrinate, flucycloxuron, flucythrinate, flufenerim, flufenoxuron, flufenprox, flumethrin, flupyrazofos, flutenzin (flufenzine), fluvalinate, fonofos, formetanate, formothion, fosmethilan, fosthiazate, fubfenprox (fluproxyfen), furathiocarb, gamma-cyhalothrin, gamma-HCH, gossyplure, grandlure, granulosis viruses, halfenprox, halofenozide, HCH, HCN-801, heptenophos, hexaflumuron, hexythiazox, hydramethylnone, hydroprene, IKA-2002, imidacloprid, imiprothrin, indoxacarb, iodofenphos, iprobenfos, isazofos, isofenphos, isoprocarb, isoxathion, ivermectin, japonilure, kadethrin, nuclear polyhedrosis viruses, kinoprene, lambda-cyhalothrin, lindane, lufenuron, malathion, mecarbam, mesulphenfos, metaldehyde, metam-sodium, methacrifos, methamidophos, Metharhizium anisopliae, Metharhizium flavoviride, methidathion, methiocarb, methomyl, methoprene, methoxychlor, methoxyfenozide, metofluthrin, metolcarb, metoxadiazone, mevinphos, milbemectin, milbernycin, MKI-245, MON-45700, monocrotophos, moxidectin, MTI-800, naled, NC-104, NC-170, NC-184, NC-194, NC-196, niclosamide, nicotine, nitenpyram, nithiazine, NNI-0001, NNI-0101, NNI-0250, NNI-9768, novaluron, noviflumuron, OK-5101, OK-5201, OK-9601, OK-9602, OK-9701, OK-9802, omethoate, oxamyl, oxydemeton-methyl, Paecilomyces fumosoroseus, parathion-methyl, parathion (-ethyl), permethrin (cis-, trans-), petroleum, PH-6045, phenothrin (1R-trans isomer), phenthoate, phorate, phosalone, phosmet, phosphamidon, phosphocarb, phoxim, piperonyl butoxide, pirimicarb, pirimiphos-methyl, pirimiphos-ethyl, potassium oleate, prallethrin, profenofos, profluthrin, promecarb, propaphos, propargite, propetamphos, propoxur, prothiofos, prothoate, protrifenbute, pymetrozine, pyraclofos, pyresmethrin, pyrethrum, pyridaben, pyridalyl, pyridaphenthion, pyridathion, pyrimidifen, pyriproxyfen, quinalphos, resmethrin, RH-5849, ribavirin, RU-12457, RU-15525, S-421, S-1833, salithion, sebufos, SI-0009, silafluofen, spinosad, spirodiclofen, spiromesifen, sulphluramid, sulphotep, sulprofos, SZI-121, tau-fluvalinate, tebufenozide, tebufenpyrad, tebupirimfos, teflubenzuron,

tetluthrin, temephos, temivinphos, terbam, terbufos, tetrachlorvinphos, tetradifon, tetramethrin, tetramethrin (1R-isomer), tetrasul, theta-cypermethrin, thiacloprid, thiamethoxam, thiapronil, thiatriphos, thiocyclam hydrogenoxalate, thiodicarb, thiofanox, thiometon, thiosultap-sodium, thuringiensin, tolfenpyrad, tralocythrin, tralomethrin, transfluthrin, triarathene, triazamate, triazophos, triazuron, trichlophenidine, trichlorfon, Trichoderma atroviride, triflumuron, trimethacarb, vamidothion, vañiliprole, verbutin, Verticillium lecanii, WL-108477, WL-40027, YI-5201, YI-5301, YI-5302, XMC, xylylcarb, ZA-3274, zeta-cypermethrin, zolaprofos, ZXI-8901, the compound 3-methylphenyl propylcarbamate (tsumacide Z), the compound 3-(5-chloro-3-pyridinyl)-8-(2,2,2-trifluoroethyl)-8-azabicyclo[3.2.1]octane-3-carbonitrile (CAS-Reg. No. 185982-80-3) and the corresponding 3-endoisomer (CAS-Reg. No. 185984-60-5) (cf. WO-96/37494, WO-98/25923), and preparations which comprise insecticidally active plant extracts, nematodes, fungi or viruses.

A mixture with other known active compounds, such as herbicides, or with fertilizers and growth regulators, safeners and/or semiochemicals is also possible.

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In addition, the compounds of the formula (I) according to the invention also have very good antimycotic activity. They have a very broad antimycotic activity spectrum in particular against dermatophytes and yeasts, moulds and diphasic fungi (for example against Candida species such as Candida albicans, Candida glabrata) and Epidermophyton floccosum, Aspergillus species such as Aspergillus niger and Aspergillus fumigatus, Trichophyton species such as Trichophyton mentagrophytes, Microsporon species such as Microsporon canis and audouinii. The list of these fungi by no means limits the mycotic spectrum which can be covered, but is only for illustration.

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The active compounds can be used as such, in the form of their formulations or the use forms prepared therefrom, such as ready-to-use solutions, suspensions, wettable powders, pastes, soluble powders, dusts and granules. Application is carried out in a customary manner, for example by watering, spraying, atomizing, broadcasting, dusting, foaming, spreading, etc. It is furthermore possible to apply the active compounds by the ultra-low volume method, or to inject the active compound preparation or the active compound itself into the soil. It is also possible to treat the seeds of the plants.

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When using the active compounds according to the invention as fungicides, the application rates can be varied within a relatively wide range, depending on the kind of application. For the treatment of parts of plants, the active compound application rates are generally between 0.1 and 10,000 g/ha, preferably between 10 and 1000 g/ha. For seed dressing, the active compound application rates are generally between 0.001 and 50 g per kilogram of seed, preferably between 0.01 and 10 g per

kilogram of seed. For the treatment of the soil, the active compound application rates are generally between 0.1 and 10,000 g/ha, preferably between 1 and 5,000 g/ha.

As already mentioned above, it is possible to treat all plants and their parts according to the invention. In a preferred embodiment, wild plant species and plant cultivars, or those obtained by conventional biological breeding, such as crossing or protoplast fusion, and parts thereof, are treated. In a further preferred embodiment, transgenic plants and plant cultivars obtained by genetic engineering, if appropriate in combination with conventional methods (Genetically Modified Organisms), and parts thereof, are treated. The term "parts" or "parts of plants" or "plant parts" has been explained above.

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Particularly preferably, plants of the plant cultivars which are in each case commercially available or in use are treated according to the invention. Plant cultivars are to be understood as meaning plants having new properties ("traits") and which have been obtained by conventional breeding, by mutagenesis or by recombinant DNA techniques. They can be cultivars, varieties, bio- or genotypes.

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Depending on the plant species or plant cultivars, their location and growth conditions (soils, climate, vegetation period, diet), the treatment according to the invention may also result in superadditive ("synergistic") effects. Thus, for example, reduced application rates and/or a widening of the activity spectrum and/or an increase in the activity of the substances and compositions which can be used according to the invention, better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest yields, better quality and/or a higher nutritional value of the harvested products, better storage stability and/or processability of the harvested products are possible which exceed the effects which were actually to be expected.

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The transgenic plants or plant cultivars (i.e. those obtained by genetic engineering) which are preferably to be treated according to the invention include all plants which, in the genetic modification, received genetic material which imparted particularly advantageous useful properties ("traits") to these plants. Examples of such properties are better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest yields, better quality and/or a higher nutritional value of the harvested products, better storage stability and/or processability of the harvested products. Further and particularly emphasized examples of such properties are a better defence of the plants against animal and microbial pests, such as against insects, mites, phytopathogenic fungi, bacteria and/or viruses, and also increased tolerance of the plants to certain herbicidally active compounds. Examples of transgenic plants which may be

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mentioned are the important crop plants, such as cereals (wheat, rice), maize, soya beans, potatoes, cotton, tobacco, oilseed rape and also fruit plants (with the fruits apples, pears, citrus fruits and grapes), and particular emphasis is given to maize, soya beans, potatoes, cotton, tobacco and oilseed rape. Traits that are emphasized are in particular increased defence of the plants against insects, arachnids, nematodes and slugs and snails by toxins formed in the plants, in particular those formed in the plants by the genetic material from Bacillus thuringiensis (for example by the genes CryIA(a), CryIA(b), CryIA(c), CryIIA, CryIIIA, CryIIIB2, Cry9c, Cry2Ab, Cry3Bb and CryIF and also combinations thereof) (hereinbelow referred to as "Bt plants"). Traits that are also particularly emphasized are the increased defence of the plants against fungi, bacteria and viruses by systemic acquired resistance (SAR), systemin, phytoalexins, elicitors and resistance genes and correspondingly expressed proteins and toxins. Traits that are furthermore particularly emphasized are the increased tolerance of the plants to certain herbicidally active compounds, for example imidazolinones, sulphonylureas, glyphosate or phosphinotricin (for example the "PAT" gene). The genes which impart the desired traits in question can also be present in combination with one another in the transgenic plants. Examples of "Bt plants" which may be mentioned are maize varieties, cotton varieties, soya bean varieties and potato varieties which are sold under the trade names YIELD GARD® (for example maize, cotton, soya beans), KnockOut® (for example maize), StarLink® (for example maize), Bollgard® (cotton), Nucoton® (cotton) and NewLeaf® (potato). Examples of herbicide-tolerant plants which may be mentioned are maize varieties, cotton varieties and soya bean varieties which are sold under the trade names Roundup Ready® (tolerance to glyphosate, for example maize, cotton, soya bean), Liberty Link® (tolerance to phosphinotricin, for example oilseed rape), IMI® (tolerance to imidazolinones) and STS® (tolerance to sulphonylureas, for example maize). Herbicide-resistant plants (plants bred in a conventional manner for herbicide tolerance) which may be mentioned also include the varieties sold under the name Clearfield® (for example maize). Of course, these statements also apply to plant cultivars which have these genetic traits or genetic traits still to be developed, and which will be developed and/or marketed in the future.

The plants listed can be treated according to the invention in a particularly advantageous manner with the compounds of the general formula (I) or the active compound mixtures according to the invention. The preferred ranges stated above for the active compounds or mixtures also apply to the treatment of these plants. Particular emphasis is given to the treatment of plants with the compounds or mixtures specifically mentioned in the present text.

The preparation and the use of the active compounds according to the invention is illustrated by the examples below.

Preparation examples

Example 1

326.5 mg (2.0 mmol) of [2-(3-methylbutyl)phenyl]amine (III-1) are added to a solution comprising 388.5 mg (2.2 mmol) of 5-fluoro-1,3-dimethyl-1H-pyrazole-4-carbonyl chloride and 0.45 ml (3.2 mmol) of triethylamine in 20 ml of tetrahydrofuran. The reaction solution is stirred at 60°C for 90 min, filtered through silica gel and concentrated. Column chromatography (cyclohexane/ethyl acetate gradient) gives 592 mg (98% of theory) of 5-fluoro-1,3-dimethyl-N-[2-(3-methylbutyl)phenyl]-1H-pyrazole-4-carboxamide of logP (pH 2.3) = 3.12.

The compounds of the formula (I) listed in Table 1 below are obtained analogously to Example 1 and in accordance with the instructions in the general descriptions of the processes.

15 <u>Table 1</u>

Ex.	R ¹	R ²	R ³	Α	logP
2	н	н	CH₃	H ₃ C N N CH ₃	3.42
3	Н	Н	CH₃	H ₃ C	4.14
4	Н	Н	Н.	CF ₃	3.90
5	Н	H	CH₃	CF ₃	4.13

Ex.	R ¹	R ²	R ³	Α	logP
6	Н	Н	Н		3.87
7	Н	Н	CH₃		4.15
8	Н	Н	H	Br	3.78
9	Н	Н	н	ō	3.76
10	н	н	н	F ₃ C N N CH ₃	3.68
11	н	Н	CH₃	F ₂ HC	3.28
12	Н	Н	CH₃	F ₃ C F	3.91
13	Н	4-CI	C₂H₅	CF ₃	4.96
14	Н	4-CI	C₂H₅		5.02
15	H	4-CI	C₂H₅	H ₃ C N N CH ₃	4.29
16	Н	Н	CH₃	F ₃ C N S	4.54

Ex.	R ¹	R ²	R ³	Α	logP
17	Н	Н	C₂H₅	H ₃ C N N CH ₃	3.68
18	н	Н	C₂H₅	F ₂ HC	3.59
19	Н	4-CI	C₂H₅	F ₂ HC	4.10
20	Н	4-F	CH₃	H ₃ C N N CH ₃	3.34
21	Н	4-F	CH₃	H ₃ C	4.14
22	Н	4-F	CH₃	F ₂ HC	3.34
23	Н	4-F	CH₃	F ₃ C N N CH ₃	3.57
24	H.	4-F	CH₃	F ₃ C N CH ₃	4.05

Preparation of starting materials of the formula (III)

Example (III-1)

A solution comprising 8.0 g (0.045 mol) of 1-(2-aminophenyl)-3-methylbutan-1-one (X-1), 6.8 g (0.135 mol) of hydrazine hydrate and 7.6 g (0.135 mol) of potassium hydroxide in 90 ml of triethylene glycol is heated at 210°C for 6 h. For work-up, water and ethyl acetate are added at room temperature. The organic phase is again washed with water, dried over magnesium sulphate and concentrated under reduced pressure. Purification by column chromatography (cyclohexane/ethyl acetate 3:1) gives 5.3 g (71.5% of theory) of [2-(3-methylbutyl)-phenyl]amine.

Example (III-2)

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3.23 g (15 mmol) of N-[2-(3,3-dimethylbut-1-ynyl)phenyl]acetamide (XIII-1) were initially charged in 40 ml of methanol. 0.5 g of palladium-on-carbon (5%) was added, and the mixture is then hydrogenated in an autoclave at a hydrogen pressure of 4 bar for 20 h. Removal of the catalyst and the solvent gave 3.1 g (94% of theory) of N-[2-(3,3-dimethylbutyl)phenyl]acetamide of logP (pH 2.3) = 2.69.

Example (III-3)

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0.5 g (2.3 mmol) of N-[2-(3,3-dimethylbutyl)phenyl]acetamide (III-2) were stirred in 20 ml of 2N hydrochloric acid at 100°C for 5 h. After cooling, the mixture was extracted 3 times with in each case 20 ml of ethyl acetate. The organic phase was removed, dried over sodium sulphate and concentrated. This gave 390 mg (29% of theory) of 2-(3,3-dimethylbutyl)phenylamine hydrochloride of logP (pH 2.3) = 2.20.

Preparation of starting materials of the formula (V)

Example (V-1)

At room temperature, 355.0 mg (2.0 mmol) of 1-(2-aminophenyl)-3-methylbutan-1-one are added to a solution comprising 388.5 mg (2.2 mmol) of 5-fluoro-1,3-dimethyl-1H-pyrazole-4-carbonyl chloride and 0.45 ml (3.2 mmol) of triethylamine in 20 ml of tetrahydrofuran. The reaction mixture is stirred at 60°C for 1.5 h, filtered through silica gel and concentrated. Column chromatography (cyclohexane/ethyl acetate: 3/1) gives 577.7 mg (1.8 mmol, 88% of theory) of N-[2-(3-methylbutyryl)phenyl-5-fluoro-1,3-dimethyl-1H-pyrazole-4-carboxamide of logP (pH = 2.3) = 3.42.

The compounds of the formula (V) listed in Table 2 below are obtained analogously to Example (V-1) and in accordance with the instructions in the general descriptions of the processes.

15 <u>Table 2</u>

Ex.	R ¹	R ²	R ³	A	logP
V-2	Н	Н	Н	H ₃ C	4.30
V-3	Н	н	Н	CF ₃	4.33
V-4	Н	Н	Н		4.53

Ex.	R ¹	R ²	R ³	Α	logP
V-5	Н	Н	Н	Br	4.35
V-6	H.	Н	Н	<u>-</u>	4.30
V-7	н	Н	Н	CH₃	4.44

Preparation of starting materials of the formula (VII)

5 Example (VII-1)

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190 mg (1.0 mmol) of 2-trifluoromethylbenzoic acid, 178 mg (0.83 mmol) of 2-(3,3-dimethyl-but-1-ynyl)phenylamine, 215 mg (1.67 mmol) of N,N-diisopropylethylamine and 583 mg (1.25 mmol) of PyBrOP in 8 ml of acetonitrile were stirred at room temperature for 4 days. 10 ml of ethyl acetate/water 1:1 were added to the mixture and the organic phase was separated off and washed with 10 ml of saturated ammonium chloride solution and subsequently with 10 ml of water. Separation, concentration and drying of the organic phase gave 950 mg of crude product. Column-chromatographic purification on silica gel 60 (petroleum ether/ethyl acetate $10:1 \rightarrow$ ethyl acetate) gave 110 mg of N-[2-(3,3-dimethylbut-1-ynyl)phenyl]-2-trifluoromethylbenzamide [logP (pH 2.3) = 4.55].

The compounds of the formula (VII) listed in Table 3 below are obtained analogously to Example (VII-1) and in accordance with the instructions in the general descriptions of the processes.

Table 3

$$\begin{array}{c|c}
O & & & \\
R^1 & & & \\
R^3 & & & \\
H_3C & & CH_3
\end{array}$$
(VII)

Ex.	R ¹	R²	R ³	Α	logP
VII-2	н	н	CH₃		4.73
VII-3	Н	н	CH₃	CI	3.75
VII-4	Н	Н	CH₃	CH ₃	4.52
VII-5	H	н	CH₃	F ₂ HC N S CH ₃	4.17

5 Preparation of starting materials of the formula (X)

Example (X-1)

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At reflux, a solution of 29.5 g (0.25 mol) of anthranilonitrile in 150 ml of tetrahydrofuran is added dropwise to a suspension comprising 18.2 g (0.75 mmol) of magnesium, 375 ml of a 2 M solution of isobutylmagnesium bromide in tetrahydrofuran and 15 ml of diethyl ether. After 5 h of heating under reflux, 100 ml of water are, at 0°C, added to the reaction mixture, and the pH is adjusted to 6 using hydrochloric acid. The organic phase is washed with water and dried over magnesium sulphate. Concentration under reduced pressure and purification on silica gel (mobile phase: petroleum

ether/acetonitrile 95:5) gives 11.0 g (25% of theory) of 1-(2-aminophenyl)-3-methylbutan-1-one of $\log P \text{ (pH 2.3)} = 2.89$.

Preparation of starting materials of the formula (XIII)

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Example (XIII-1)

Under argon, 25.7 g (120 mmol) of ortho-bromoacetanilide, 5.05 g (7.2 mmol) of bis(triphenyl-phosphine)palladium(II) chloride and 1.37 g (7.2 mmol) of copper(I) iodide were initially charged in 450 ml of triethylamine. At room temperature, 17.8 g (180 mmol) of 3,3-dimethyl-1-butyne were then added dropwise over a period of 10 min, and the mixture is stirred at 50° C for 5 h. The reaction mixture was poured into 21 of water and extracted 3 times with in each case 250 ml of diethyl ether, and the extracts were dried over sodium sulphate and concentrated. Column-chromatographic purification on silica gel 60 with methylene chloride gave 25.9 g of N-[2-(3,3-dimethylbut-1-ynyl)-phenyl]acetamide of logP (pH 2.3) = 3.03.

The determination of the logP values given in preparation examples and tables above is carried out according to EEC-Directive 79/831 Annex V.A8 by HPLC (High Performance Liquid Chromatography) on a reversed phase column (C 18). Temperature: 43°C.

- Determination is carried out in the acidic range at pH 2.3 using the mobile phases 0.1% aqueous phosphoric acid and acetonitrile; linear gradient from 10% acetonitrile to 90% acetonitrile.
 - Calibration is carried out using unbranched alkan-2-ones (of 3 to 16 carbon atoms) whose logP values are known (determination of the logP values by retention times using linear interpolation between two successive alkanones).
- The lambda-max values were determined in the maxima of the chromatographic signals using the UV spectra from 200 nm to 400 nm.

Use examples:

Example A

5 Podosphaera test (apple) / protective

Solvents:

24.5 parts by weight of acetone

24.5 parts by weight of dimethylacetamide

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

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To produce a suitable preparation of active compound, 1 part by weight of compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

- To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rates. After the spray coating has dried on, the plants are inoculated with an aqueous spore suspension of the apple mildew pathogen Podosphaera leucotricha. The plants are then placed in a greenhouse at about 23°C and a relative atmospheric humidity of about 70%.
- Evaluation is carried out 10 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

<u>Table A</u>

Podosphaera test (apple) / protective

Active compound according to the invention	Application rate of active compound in g/ha	Efficacy in %
H ₃ C O N H ₃ C CH ₃	100	100
H ₃ C CH ₃ CH ₃	100	100
H ₃ C O N CH ₃ CH ₃	100	99
H ₃ C CH ₃	100	97
H ₃ C O N CH ₃ CH ₃	100	99
H ₃ C CH ₃ CH ₃	100	100

Podosphaera test (apple) / protective

Active compound according to the invention	Application rate of active compound in g/ha	Efficacy in %
H ₃ C CH ₃	100	100
H ₃ C CH ₃	100	100

Example B

Venturia test (apple) / protective

5 Solvents:

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24.5 parts by weight of acetone

24.5 parts by weight of dimethylacetamide

Emulsifier:

part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rates. After the spray coating has dried on, the plants are inoculated with an aqueous conidia suspension of the apple scab pathogen Venturia inaequalis and then remain in an incubation cabin at about 20°C and 100% relative atmospheric humidity for 1 day.

The plants are then placed in a greenhouse at about 21°C and a relative atmospheric humidity of about 90%.

<u>Table B</u>

Venturia test (apple) / protective

Active compound according to the invention	Application rate of active compound in g/ha	Efficacy in %
H ₃ C O N H ₃ C CH ₃	100	96
H ₃ C O CH ₃ H ₃ C CH ₃	100	100
H ₃ C O CH ₃ CH ₃	100	99
H ₃ C CH ₃	100	97
H ₃ C O N CH ₃ CH ₃ C CH ₃	100	100
H ₃ C CH ₃ CH ₃	100	100

Venturia test (apple) / protective

Active compound according to the invention	Application rate of active compound in g/ha	Efficacy in %
H ₃ C CH ₃	100	100
H ₃ C CH ₃ CH ₃	100	100
H ₃ C CH ₃	100	100

Example C

Botrytis test (bean) / protective

5 Solvents:

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24.5 parts by weight of acetone

24.5 parts by weight of dimethylacetamide

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rates. After the spray coating has dried on, 2 small pieces of agar colonized by Botrytis cinerea are placed onto each leaf. The inoculated plants are placed in a dark chamber at about 20°C and 100% relative atmospheric humidity.

Two days after the inoculation, the size of the infected areas on the leaves is evaluated. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

<u>Table C</u>

Botrytis test (bean) / protective

Active ingredient according to the invention	Application rate of active compound in g/ha	Efficacy in %
H ₃ C O N H ₃ C CH ₃	500	84
H ₃ C O CH ₃ CH ₃	500	100
H ₃ C O CH ₃ CH ₃	500	100
H ₃ C CH ₃	500	87
H ₃ C O CH ₃ CH ₃	500	100
H ₃ C CH ₃	500	100

Botrytis test (bean) / protective

Active ingredient according to the invention	Application rate of active compound in g/ha	Efficacy in %
F ₂ HC O CH ₃ H ₃ C CH ₃	500	100
H ₃ C CH ₃ CH ₃	500	100
F ₂ HC O F CH ₃ CH ₃	500	100

Example D

Puccinia test (wheat) / curative

5 Solvent:

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50 parts by weight of N,N-dimethylacetamide

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for curative activity, young plants are sprayed with a conidia suspension of Puccinia recondita. The plants remain in an incubation cabin at 20°C and 100% relative atmospheric humidity for 48 hours. The plants are then sprayed with the preparation of active compound at the stated application rates.

The plants are then placed in a greenhouse at a temperature of about 20°C and a relative atmospheric humidity of 80% to promote the development of rust pustules.

Evaluation is carried out 8 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

Table D

Puccinia test (wheat) / curative

Active ingredient according to the invention	Application rate of active ingredient in g/ha	Efficacy in %
H ₃ C O N H H ₃ C CH ₃	500	. 100
H ₃ C O N CH ₃ CH ₃	500	100
H ₃ C O CH ₃ CH ₃	500	100
CF ₃ O H ₃ C CH ₃	500	100
H ₃ C CH ₃	500	100
H ₃ C O CH ₃ CH ₃	500	100

Puccinia test (wheat) / curative

Active ingredient according to the invention	Application rate of active ingredient in g/ha	Efficacy in %
F ₂ HC O N H ₃ C CH ₃ CH ₃	500	100
F ₂ HC O N H CH ₃ H ₃ C CH ₃	500	100
F ₂ HC O F F CH ₃ CH ₃ CH ₃	500	100

Example E

Sphaerotheca test (cucumber) / protective

5 Solvent:

10

15

49 parts by weight of N, N-dimethylformamide

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young cucumber plants are sprayed with the preparation of active compound at the stated application rates. One day after the treatment, the plants are inoculated with a spore suspension of Sphaerotheca fuliginea. The plants are then placed in a greenhouse at 70% relative atmospheric humidity and a temperature of 23°C.

Evaluation is carried out 7 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

<u>Table E</u> **Sphaerotheca test (cucumber) / protective**

Active ingredient according to the invention	Application rate of active compound in g/ha	Efficacy in %
H ₃ C O N H ₃ C CH ₃	750	100
H ₃ C CH ₃ CH ₃	750	100
CF ₃ O N H ₃ C CH ₃	750	100
N CH ₃ CH ₃	750	100